

# Order-by-Disorder Near Criticality in $XY$ Pyrochlore Magnets

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We consider a system of spins on the sites of a three-dimensional pyrochlore lattice of corner-sharing tetrahedra interacting with a predominant effective  $xy$  exchange. In particular, we investigate the selection of a long-range ordered state with broken discrete symmetry induced by thermal fluctuations near the critical region. At the standard mean-field theory (s-MFT) level, in a region of the parameter space of this Hamiltonian that we refer to as  $\Gamma_5$  region, the ordered state possesses an accidental  $U(1)$  degeneracy. In this paper, we show that fluctuations beyond s-MFT lift this degeneracy by selecting one of two states (so-called  $\psi_2$  and  $\psi_3$ ) from the degenerate manifold, thus exposing a certain form of order-by-disorder (ObD). We analytically explore this selection at the microscopic level and close to criticality by elaborating upon and using an extension of the so-called TAP method, originally developed by Thouless, Anderson and Palmer to study the effect of fluctuations in spin glasses. We also use a single-tetrahedron cluster-mean-field theory (c-MFT) to explore over what minimal length scale fluctuations can lift the degeneracy. We find the phase diagrams obtained by these two methods to be somewhat different since c-MFT only includes the shortest-range fluctuations. General symmetry arguments used to construct a Ginzburg-Landau theory to lowest order in the order parameters predict that a weak magnetic moment,  $m_z$ , along the local  $\langle 111 \rangle$  ( $\hat{z}$ ) direction is generically induced for a system ordering into a  $\psi_2$  state, but not so for  $\psi_3$  ordering. Both E-TAP and c-MFT calculations confirm this weak fluctuation-induced  $m_z$  moment. Using a Ginzburg-Landau theory, we discuss the phenomenology of multiple phase transitions below the paramagnetic phase transition and within the  $\Gamma_5$  long-range ordered phase.

## I. INTRODUCTION

In the study of condensed matter systems, mean-field theory<sup>1</sup> is often the simplest starting point to obtain a qualitative understanding of the essential physics at play prior to carrying out a more sophisticated analysis. A standard mean-field theory (s-MFT) replaces the many-body problem with a simpler problem of a one-body system interacting with an averaged field produced by the rest of the interacting particles. In systems with competing or frustrated interactions, s-MFT may yield a number of states with a degenerate minimum free energy below the mean-field critical temperature,  $T_c^{\text{MF}}$ .<sup>2,3</sup> If these degeneracies are accidental, that is not imposed by exact symmetries of the Hamiltonian, they may be lifted by the effects of thermal<sup>4</sup> or quantum<sup>5</sup> fluctuations, a phenomenon known as order-by-disorder (ObD).<sup>4-8</sup>

The concept of ObD was originally proposed by Villain and collaborators as the ordered state selection mechanism for a two-dimensional frustrated Ising model on the domino lattice.<sup>4</sup> Since this seminal work, ObD has been theoretically identified and discussed for many highly frustrated magnetic models.<sup>6-18</sup> These systems generically possess an exponentially ( $\exp[N^\alpha]$ ) large number of classical ground-states (here  $N$  is the number of spins in the system and  $\alpha \leq 1$ ). As a result, highly frustrated magnetic systems are intrinsically very sensitive to fluctuations or energetic perturbations. In the context of experimental studies of real materials, it is difficult to distinguish a selection of an ordered state via fluctuations from one that would arise from energetic perturbations beyond the set of interactions considered in a restricted theoretical model. Consequently, undisputed examples of ObD in experiments

have remained scarce.<sup>19-21</sup>

Quite recently, following an original proposal going back ten years,<sup>22,23</sup> and building on an earlier study,<sup>24</sup> several papers<sup>15-18</sup> have put forward compelling arguments for ObD being responsible for the experimentally observed long-range order in the insulating rare-earth pyrochlore oxide<sup>25</sup>  $\text{Er}_2\text{Ti}_2\text{O}_7$ . In this compound,  $\text{Er}^{3+}$  is magnetic and  $\text{Ti}^{4+}$  is not. The key observation in those works is that the accidentally degenerate classical ground states are related by operations with a  $U(1)$  symmetry.<sup>16</sup> This set of classically degenerate states form the so-called  $\Gamma_5$  manifold.<sup>26</sup> For a range of interaction parameters of the most general symmetry-allowed bilinear nearest-neighbor pseudospin  $1/2$  exchange-like Hamiltonian (referred to as  $\mathcal{H}$  in Eq. (1a) below) on the pyrochlore lattice of corner-sharing tetrahedra (see Fig. 3), the  $U(1)$  degeneracy is exact at the s-MFT level as long as the cubic symmetry of the system remains intact. As a shorthand, we henceforth refer to this region of exchange parameter space as the  $\Gamma_5$  region, as referred to in the Abstract. Reference [16] showed that the  $U(1)$  symmetry is robust against a wide variety of perturbations added to  $\mathcal{H}$  and argued that essentially only fluctuations can efficiently lift the degeneracy in  $\text{Er}_2\text{Ti}_2\text{O}_7$  when considering bilinear anisotropic interactions of arbitrary range between the pseudospins.<sup>27</sup> Similar arguments were made in Ref. [15]. In this compound, a particular long-range ordered state, the  $\psi_2$  state (see Fig. (1)),<sup>22,23,26,28-31</sup> is selected. Considering  $\mathcal{H}$  on the pyrochlore lattice, Wong *et al.*<sup>17</sup> and Yan *et al.*<sup>32</sup> studied the effect of quantum<sup>17,32</sup> and thermal<sup>32</sup> fluctuations and established a general phase diagram for this Hamiltonian at  $T = 0^+$ .

The investigations reported in Refs. [14–17, and 32] fo-

cused on identifying the mechanism of *ground state* selection by taking into account the harmonic quantum<sup>14–17,32</sup> or classical<sup>14,32</sup> spin fluctuations about a classical long-range ordered state. On the other hand, the problem of state selection at temperatures near the critical transition temperature to the paramagnetic phase has received significantly less attention. Although there have been numerical studies of ObD selection of the  $\psi_2$  state at  $T \lesssim T_c$ ,<sup>14,15,22,23,32–34</sup> or upon approaching  $T_c$  from above,<sup>18</sup> an analytical study specifying the role of the individual microscopic anisotropic spin-spin interactions in the ObD mechanism at  $T \approx T_c$  has, to the best of our knowledge, not yet been carried out. The problem of selection at  $T \lesssim T_c$  is not only of relevance to the phenomenology of  $\text{Er}_2\text{Ti}_2\text{O}_7$  or other pyrochlore magnetic compounds,<sup>25</sup> but it is of considerable interest for all highly frustrated magnetic systems proposed to display an ObD mechanism.

There are situations where a sort of ObD occurs near  $T_c$  which differ from the textbook cases<sup>4–6,8</sup> where the state selection near  $T = 0^+$ , proceeding either via thermal or quantum selection, is leveraged upon all the way to the long-range ordered state selected at  $T_c$ . For example, the long-range ordered state selected by ObD can in principle be different for the  $T = 0^+$  and  $T \lesssim T_c$  regimes. This occurs, for example, for classical Heisenberg spins on the pyrochlore lattice interacting via nearest-neighbor antiferromagnetic exchange and indirect Dzyaloshinskii-Moriya interaction.<sup>34,35</sup> In another class of problems, different competing long-range ordered states may have the same free-energy at the s-MFT level only over a finite temperature interval,  $T^* \leq T \leq T_c$  with a transition to a *non-degenerate* classical ground state at  $T^*$ . There, thermal fluctuation corrections to s-MFT can select one of the competing states over the  $T^* \leq T \leq T_c$  window. This is what is predicted to occur in the multiple- $k$  state selection in the  $\text{Gd}_2\text{Ti}_2\text{O}_7$  pyrochlore antiferromagnetic between 0.7 K and 1.0 K.<sup>36</sup> Perhaps the most exotic cases arise when a relic of ObD occurs at the critical temperature, while the classical ground state is *not* degenerate for the Hamiltonian considered, but would be for a closely related Hamiltonian minus some degeneracy-lifting weak energetic perturbations.<sup>14,37,38</sup> At the phenomenological Ginzburg-Landau level description, these cases are not paradoxical. They only become so when one is trying to ascribe a microscopic description to the physics at stake and the origin for, at least, one further equilibrium thermodynamic phase transition at some temperature below the paramagnetic transition at  $T_c$ . Finally, and generally speaking, one may ask whether a discussion of ObD at  $T = 0^+$  is of pragmatic usefulness given that most experiments for which ObD may pertain typically proceed by cooling a material from a paramagnetic disordered phase to an ordered phase and not going below a certain baseline nonzero temperature constrained by the experimental set-up. This broader context provides the motivation for our work which aims to go beyond the sole consideration of a phenomenological Ginzburg-Landau theory that contains all terms, relevant or not (in the renormalization-group sense), allowed by symmetry and to expose how the different competing microscopic interactions participate to the degeneracy-lifting near the transition at  $T_c$ .

In the temperature regime near a phase transition, the har-

monic approximation describing low-energy excitations usually employed in theoretical discussions of ObD<sup>5–8</sup> at  $T = 0^+$  is not physically justified since large fluctuations typically accompany the phase transition to the paramagnetic state. From a fundamental viewpoint, it is thus highly desirable to study the role of fluctuations beyond s-MFT using a different method that can be applied at temperatures close to the critical region. A possible route to tackle this problem was paved by Thouless, Anderson and Palmer (TAP) in their study of the effect of fluctuations in spin glasses.<sup>39</sup> For the case of the Ising spin glass model, Plefka showed that the TAP correction can be systematically derived using a perturbative expansion,<sup>40</sup> an approach that was later generalized by Georges and Yedidia who calculated higher order terms in the Plefka's perturbative expansion.<sup>41</sup> This approach, which we refer to as extended TAP (E-TAP), consists of a high-temperature expansion in  $\beta \equiv 1/k_B T$  about the s-MFT solution. It captures corrections to the s-MFT free-energy by including fluctuations in the form of on-site linear and nonlinear susceptibilities. A second approach allowing one to go beyond s-MFT is the cluster mean-field theory (c-MFT).<sup>42,43</sup> This approach treats short-range fluctuations within a finite cluster exactly while taking into account the inter-cluster interactions in a mean-field fashion.

In the present work, we focus on the problem of ObD selection near criticality in a spin model on the pyrochlore lattice with predominant  $xy$  interactions using the E-TAP method. We have recently used this method to investigate the problem of partial multiple- $k$  order in pyrochlore magnets.<sup>36</sup> In this paper, we also employ c-MFT. We concentrate on the  $\Gamma_5$  manifold for the  $T \lesssim T_c^{\text{MF}}$  regime since  $\Gamma_5$  is not only an interesting theoretical playground according to recent investigations,<sup>14–17,31–33,44</sup> it is also of potential relevance to real pyrochlore materials, such as  $\text{Er}_2\text{Ti}_2\text{O}_7$ , proposed to display an ObD mechanism.<sup>15,16,22,30</sup> In addition, the interesting case of distinct ObD selection at  $T = 0^+$  and  $T_c$ , reported in a pyrochlore system with anisotropic spin-spin coupling,<sup>34</sup> further motivates us in investigating the role of the various spin interactions in the state selection at  $T \lesssim T_c$  in a general model of interacting spins on the pyrochlore lattice.

The rest of the paper is organized as follows. In Section II, we present the bilinear nearest-neighbor spin model defined by the Hamiltonian  $\mathcal{H}$  of Eq. (1a) on the pyrochlore lattice and discuss its symmetries. Focusing on the  $\Gamma_5$  manifold, we present a Ginzburg-Landau (GL) symmetry analysis to specify the general form of the lowest-order anisotropic terms allowed in the GL free-energy ( $\mathcal{F}_{\text{GL}}$ ) that can lift the accidental  $U(1)$  degeneracy found in s-MFT. We show that the fluctuation correction terms to the s-MFT free-energy select either the  $\psi_2$  or the  $\psi_3$  long-range ordered state of the  $\Gamma_5$  manifold (see Fig. 1 and Appendix A for definition) as well as induce a weak moment  $m_z$  along the local [111] direction for the  $\psi_2$  state. In Section III A, we present the E-TAP method and determine the phase boundary at  $T_c$  between the  $\psi_2$  and  $\psi_3$  states in the space of anisotropic spin-spin coupling constants. We investigate in Section III B how short-ranged fluctuations acting on the length-scale of a single-tetrahedron can lead to ObD when incorporated in the self-consistent scheme

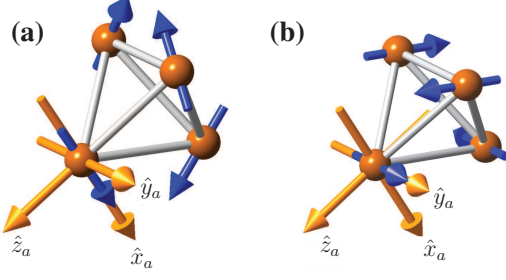


FIG. 1. (Color Online). Spin configurations of the (a) non-coplanar  $\psi_2$  state, as observed in  $\text{Er}_2\text{Ti}_2\text{O}_7$ ,<sup>26</sup> and the (b) coplanar  $\psi_3$  state on a single tetrahedron. The pattern is the same on all tetrahedra in a pyrochlore lattice which is a face-centered cubic lattice with a tetrahedron basis and these two states, which belong to the  $\Gamma_5$  manifold, are thus said to have a  $\mathbf{k} = 0$  ordering wave vector. The spin (in blue) at the  $a$  sublattice points along (a) the  $\hat{x}_a$  axis of the local  $[111]$  reference frame for the  $\psi_2$  state and along (b) the  $\hat{y}_a$  axis of the same local frame for the  $\psi_3$  state. The local  $\hat{z}_a$  axis at sublattice  $a$  points along the local cubic  $[111]$  axis at that site such that the local  $\hat{x}_a, \hat{y}_a, \hat{z}_a$  triad of orthogonal unit vectors (orange arrows) fulfills  $\hat{x}_a \times \hat{y}_a = \hat{z}_a$ . As illustrated in panels (a) and (b), the local  $\hat{z}_a$  axis for sublattice  $a$  points directly out of the tetrahedron primitive basis cell. See Appendix A for more details.

of c-MFT. The c-MFT method also allows us to explore semi-quantitatively the role of nonzero effective Ising exchange ( $J_{zz}$ ) on the selection at  $T_c$ . In Section IV, we briefly discuss the possibility of multiple phase transitions in  $xy$  pyrochlore magnets as temperature, for example, is varied. Finally, we close with a discussion in Section V. A number of appendices are provided to assist the reader with the technical details of the E-TAP calculations. In Appendix A, we detail the spin configurations of the  $\psi_2$  and  $\psi_3$  states. In Appendix B, we analyze the symmetry properties of the  $\psi_2$  and the  $\psi_3$  states to show that a fluctuation-induced local  $m_z$  moment is only compatible with a  $\psi_2$  long-range ordered state. We provide in Appendix C the details of the E-TAP calculations and the diagrammatic approach employed.

## II. MODEL

We consider the following Hamiltonian on the pyrochlore lattice:<sup>16,45</sup>

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_1 \quad (1a)$$

$$\mathcal{H}_0 = \sum_{\langle ij \rangle} J_{zz} S_i^z S_j^z - J_{\pm} (S_i^+ S_j^- + S_i^- S_j^+) \quad (1b)$$

$$\begin{aligned} \mathcal{H}_1 = \sum_{\langle ij \rangle} & J_{\pm\pm} (S_i^+ S_j^+ \gamma_{ij} + S_i^- S_j^- \gamma_{ij}^*) \\ & + J_{z\pm} \{ S_i^z (\zeta_{ij} S_j^+ + \zeta_{ij}^* S_j^-) + i \leftrightarrow j \} \end{aligned} \quad (1c)$$

where  $S_i^{\pm} \equiv S_i^x \pm i S_i^y$  and  $S_i^{\mu}$ , with  $\mu = z, +, -$ , is defined in the local  $[111]$  coordinate frame<sup>45</sup> attached to each of the

four pyrochlore sublattices (see Fig. 1).  $S_i$  can be treated classically as a 3-component vector or quantum mechanically as an operator such as a  $S_i = 1/2$  pseudospin. In the context of magnetic rare-earth pyrochlores,  $S_i$  would represent either the total angular momentum  $\mathbf{J}$  within a simplified model of  $\mathbf{J} - \mathbf{J}$  coupling,<sup>31,46–48</sup> or a pseudospin  $1/2$  describing the single-ion ground state doublet.<sup>15,16,25,45–49</sup>

As we are foremost interested in the selection of classical ordered phases at  $0 \ll T \lesssim T_c$ , we shall treat  $S_i$  generally classically with  $|S_i| = 1/2$  for all  $i$ . However, in Section IIIB, where we use the c-MFT method, we consider  $S_i = 1/2$  quantum mechanically, mostly for computational ease. In Eq. (1),  $J_{zz}, J_{\pm}, J_{\pm\pm}$  and  $J_{z\pm}$  are the four symmetry-allowed independent nearest-neighbor exchange parameters, while  $\zeta_{ij} = -\gamma_{ij}^*$  are bond-dependent phases on a single tetrahedron defined in Refs. [16, 45, and 49].<sup>50</sup>

It is important for the discussion that follows to split  $\mathcal{H}$  into the two terms  $\mathcal{H}_0$  and  $\mathcal{H}_1$  in Eq. (1a) and consider the symmetry properties of each term.  $\mathcal{H}_0$  has a  $U(1)$  symmetry: it is invariant under a rotation of  $S_i$  by an arbitrary global angle about the local  $\langle 111 \rangle$  axes. On the other hand, the  $\mathcal{H}_1$  term in Eq. (1a) is only invariant under rotations of  $S_i$  by  $\frac{2\pi}{3}$ , reducing the symmetry of  $\mathcal{H}$  to  $Z_6$  ( $C_3[111] \times Z_2$ ).

We consider a s-MFT treatment of the Hamiltonian of Eq. (1) which orders in a state with a  $\mathbf{k} = 0$  ordering wave vector in a certain region of the parameter space centered around a dominant  $J_{\pm}$  ( $\Gamma_5$  region). In this state, all the magnetic moments in the system lie predominantly perpendicular to the local  $[111]$  direction and point in the same direction in the local coordinate system (see Fig. 1). These spin configurations define the  $\Gamma_5$  manifold.<sup>26</sup> As a result,  $\mathbf{m}_i \equiv \langle S_i \rangle$ , the on-site magnetization,<sup>51</sup> and  $\phi_i \equiv \tan^{-1}(m_i^y/m_i^x)$ , the azimuthal angle expressed in the local  $[111]$  coordinate system, are independent of the lattice site index  $i$ . We henceforth drop the index  $i$  of  $\phi_i$  and  $\mathbf{m}_i$  for these  $\mathbf{k} = 0$  spin configurations.

As first noted in Ref. [16], in the  $\Gamma_5$  region, the s-MFT free-energy is *independent* of  $\phi$  and the system displays an accidental  $U(1)$  symmetry within such a s-MFT description. However, since  $\mathcal{H}$  only has a global  $Z_6$  symmetry, we expect that in a treatment of the problem that goes beyond s-MFT, this “artificial”  $U(1)$  symmetry to be reduced to a  $Z_6$  symmetry in the paramagnetic phase which gets spontaneously broken in the ordered phase. In this context, we note that a high-temperature series expansion of the quantum model (1a), with values  $\{J_{\pm}, J_{\pm\pm}, J_{z\pm}, J_{zz}\}$  appropriate for  $\text{Er}_2\text{Ti}_2\text{O}_7$ ,<sup>16</sup> shows explicitly that such a  $Z_6$  anisotropy develops in the paramagnetic phase upon approaching  $T_c$  from above.<sup>18</sup> Also, a recent Monte Carlo study has investigated the critical behavior of a classical version of this model.<sup>33</sup> We now present a Ginzburg-Landau (GL) symmetry analysis that allows one to anticipate the form of the lowest order fluctuation corrections to s-MFT that lift the  $U(1)$  degeneracy.



### A. Ginzburg-Landau (GL) symmetry analysis

We start by defining the complex variable

$$m_{xy} \equiv \sqrt{m_x^2 + m_y^2} \exp(i\phi), \quad (2)$$

which corresponds to the magnitude and direction of the on-site magnetization  $\mathbf{m} = \langle \mathbf{S}_i \rangle$  in the local  $xy$  plane.

For simplicity, we first consider a strictly local- $xy$  state and assume  $m_z = 0$ . The allowed terms in the Ginzburg-Landau (GL) free-energy can only be of even powers in  $m_{xy}$  due to time-reversal symmetry,  $\tau$ . On the other hand, the rotation by  $2\pi/3$  about the cubic  $\langle 111 \rangle$  axes, or  $C_3$  symmetry, forbids the existence of terms of the form  $m_{xy}^n + (m_{xy}^*)^n$  unless  $n$  is a multiple of three. The resulting terms have the ability to lift the accidental  $U(1)$  degeneracy of the  $\Gamma_5$  manifold since they introduce a dependence of the GL free-energy on the azimuthal angle  $\phi$ , the orientation of  $\mathbf{m}$  in the local  $xy$  plane (see Eq. (2)). Considering the effect of  $C_3$  and  $\tau$  together, assuming strictly  $xy$  order ( $m_z = 0$ ), the lowest order term that breaks the  $U(1)$  symmetry must therefore have  $n = 6$  and be of the form

$$\eta_6(T)[m_{xy}^6 + (m_{xy}^*)^6], \quad (3)$$

where the anisotropy strength  $\eta_6$  depends on temperature,  $T$ . We note that such a sixth order term is dangerously irrelevant for the 3-dimensional  $xy$  universality class.<sup>52</sup> This means that while it does not affect the critical properties of the system,<sup>33,53</sup> this term plays a crucial role in the selection of a specific long-range ordered state by lifting the  $U(1)$  s-MFT degeneracy at  $T \lesssim T_c$ . Higher order terms  $f_{6n} \sim |m_{xy}|^{6n}$  ( $n > 1$ ) are also generated by fluctuations, but are even more irrelevant than  $f_6$  at  $T_c$ . As an example, we discuss in Section IV the effects of competing  $f_6$  and  $f_{12}$  terms at  $T < T_c$  as the amplitude  $|m_{xy}|$  grows upon cooling below  $T_c$ .

If we now include the  $z$  component of the order parameter, i.e.  $m_z \neq 0$ , a  $U(1)$  symmetry-breaking term arises at fourth order in the components of  $\mathbf{m}$  in the GL free-energy. Such a term was only recently noted in a numerical study<sup>31</sup> but whose microscopic and symmetry origins was not discussed. Again, based on the combined effect of the  $C_3$  and  $\tau$  symmetry operations, the degeneracy-lifting fourth order term in the GL free-energy has the form

$$\omega(T)m_z[m_{xy}^3 + (m_{xy}^*)^3], \quad (4)$$

where, here again, the anisotropic coupling  $\omega$  depends on  $T$ . Together, Eqs. (3, 4) identify the form of the two lowest order terms in the GL free-energy capable of lifting the degeneracy within the  $\Gamma_5$  manifold.

It turns out that the sixth order term of Eq. (3) and the fourth order term of Eq. (4) have the same net effect in lifting the degeneracy beyond s-MFT as can be shown by combining them into a single term in the GL free-energy,  $\mathcal{F}_{\text{GL}}$ . Consider first the quadratic terms in  $\mathcal{F}_{\text{GL}}$  of the form  $r_0(m_x^2 + m_y^2) + r_1 m_z^2$ . Here  $r_0$  and  $r_1$  are chosen to be different to emphasize the distinct criticality for the  $xy$  and  $z$  components of the order

parameter  $\mathbf{m}$ . Next, taking into account the terms of Eqs. (3, 4), we have:

$$\begin{aligned} \mathcal{F}_{\text{GL}} = & r_0(m_x^2 + m_y^2) + r_1 m_z^2 + \omega m_z [m_{xy}^3 + (m_{xy}^*)^3] \\ & + \eta_6 [m_{xy}^6 + (m_{xy}^*)^6] + \mathcal{F}_{\text{GL}}^{(4)}(m_{xy}, m_z) \\ & + \mathcal{F}_{\text{GL}}^{(6)}(m_{xy}, m_z) + \dots \end{aligned} \quad (5)$$

This can be rewritten as

$$\begin{aligned} \mathcal{F}_{\text{GL}} = & r_0(m_x^2 + m_y^2) + r_1(m_z + \frac{\omega |m_{xy}|^3 \cos(3\phi)}{r_1})^2 \\ & - \frac{\omega^2 |m_{xy}|^6}{2r_1} + (2\eta_6 - \frac{\omega^2}{2r_1}) |m_{xy}|^6 \cos(6\phi) \\ & + \mathcal{F}_{\text{GL}}^{(4)}(m_{xy}, m_z) + \mathcal{F}_{\text{GL}}^{(6)}(m_{xy}, m_z) + \dots, \end{aligned} \quad (6)$$

where we used Eq. (2) to go from Eq. (5) to Eq. (6). In Eqs. (5,6),  $\mathcal{F}_{\text{GL}}^{(4)}$  and  $\mathcal{F}_{\text{GL}}^{(6)}$  are fourth and sixth order terms and  $\dots$  represents terms that are of higher order in the components of  $\mathbf{m}$ . We take  $r_1 > 0$  to enforce no criticality for the  $m_z$  component of  $\mathbf{m}$ . Upon minimizing Eq. (6) with respect to  $m_z$ , we obtain:

$$m_z = -\omega |m_{xy}|^3 \cos(3\phi) / r_1. \quad (7)$$

Hence, after having minimized  $\mathcal{F}_{\text{GL}}$  with respect to  $m_z$ , we have

$$\delta \mathcal{F}_{\text{GL}}(\phi) \equiv 2\bar{\eta}_6 |m_{xy}|^6 \cos(6\phi), \quad (8)$$

with

$$2\bar{\eta}_6 \equiv (2\eta_6 - \omega^2 / 2r_1), \quad (9)$$

and where  $\delta \mathcal{F}_{\text{GL}}(\phi)$  is the anisotropic part of  $\mathcal{F}_{\text{GL}}$ , the  $U(1)$  degeneracy-lifting term on which we focus. The minimum of  $\delta \mathcal{F}_{\text{GL}}$  is either  $\cos(6\phi) = \pm 1$  depending on the sign of  $\bar{\eta}_6$ . This happens for  $\phi = n\pi/3$  or  $\phi = (2n+1)\pi/6$ , with  $n = 0, 1, \dots, 5$  with these two sets of angles corresponding, respectively, to the  $\psi_2$  and the  $\psi_3$  states (see Fig. 1). So, when  $\bar{\eta}_6 < 0$  ( $\bar{\eta}_6 > 0$ ), the minimum free-energy state is  $\psi_2$  ( $\psi_3$ ). The boundary between  $\psi_2$  and  $\psi_3$  is thus determined by the real roots of  $\bar{\eta}_6$ . A similar discussion is invoked in Ref. [17] to describe the zero temperature  $\psi_2$ - $\psi_3$  phase boundaries arising from quantum ObD.

We note that the fluctuation corrections to s-MFT induce a local  $m_z$  moment to the minimum free-energy state which was found to be strictly ordered in the local  $xy$  plane at the s-MFT level.<sup>54</sup> Since this moment is proportional to  $\cos(3\phi)$ , only the  $\psi_2$  state can have a nonzero  $m_z$ . This result is also expected based solely on symmetry considerations for the  $\psi_2$  and  $\psi_3$  states (see Appendix B). The induced moment is, however, small just below  $T_c$  since it is proportional to  $|m_{xy}|^3$  and inversely proportional to  $r_1$  which remains firmly positive away from spontaneous Ising criticality at  $r_1 = 0$ .

At the phenomenological GL level, Eq. (8) is the final result demonstrating how the  $U(1)$  degeneracy at the s-MFT level may be lifted by the anisotropic terms of Eqs. (3,4). In this work, however, we are rather interested in exposing how the coefficient of the symmetry-breaking terms in the free-energy

of Eq. (6),  $\bar{\eta}_6$ , can be determined *at a microscopic level* when going beyond a s-MFT description. Specifically, we wish to explore the leading dependence of  $\bar{\eta}_6$  upon the  $J_{\pm\pm}$ ,  $J_{z\pm}$  and  $J_{zz}$  anisotropic exchange couplings near  $T_c$ .

### III. METHODS AND RESULTS

In Section III A, we use the E-TAP method to compute the phase boundary between the  $\psi_2$  and  $\psi_3$  states determined by the function  $\bar{\eta}_6$  in Eq. (8) for  $T \lesssim T_c^{\text{MF}}$ . In this calculation, we consider for simplicity  $J_{zz} = 0$  in Eq. (1).<sup>55</sup> In Section III B, we conduct a complementary numerical study of fluctuation corrections to the s-MFT using cluster-MFT (c-MFT) which allows us to explore the effect of fluctuations at the level of one tetrahedron for both  $J_{zz} = 0$  and  $J_{zz} \neq 0$ . Since we are essentially interested in the  $\Gamma_5$  region where the dominant interaction is  $J_{\pm}$  in Eq. (1), we express the rest of the couplings in units of  $J_{\pm}$  and we henceforth denote the scaled (perturbative) interactions with lower case letters, i.e.  $j_{\pm\pm} \equiv J_{\pm\pm}/J_{\pm}$  and  $j_{z\pm} \equiv J_{z\pm}/J_{\pm}$ .

#### A. Extended-TAP Method (E-TAP)

##### 1. Method

In a magnetic system with static magnetic moments, a given moment is subject to a local field due to its neighbors. In a s-MFT treatment, the moment affects its own local field indirectly; this is an artifact of s-MFT. The so-called Onsager reaction field introduces a term in the s-MFT free-energy that cancels this unphysical effect to leading order. As was shown by Thouless, Anderson and Palmer (TAP),<sup>39</sup> this reaction-field correction is particularly important in setting up a proper mean-field theory description of spin glasses with infinite-range interactions.<sup>56</sup> Here, we present an extended version of the TAP method (E-TAP) first developed by Georges and Yedidia<sup>41</sup> for Ising spin glasses and which includes the lowest order fluctuation corrections originally calculated by TAP as well as those beyond.

To proceed, we must first modify the E-TAP procedure of Ref. [41] to study the case of 3-component classical spins with anisotropic exchange interactions. In the E-TAP method, nonzero on-site fluctuations i.e.  $\langle S_i^\alpha S_i^\beta \rangle \neq \langle S_i^\alpha \rangle \langle S_i^\beta \rangle$ ,<sup>41</sup> are taken into account via a high-temperature expansion (small  $\beta$ ) of a Gibbs free-energy:

$$G = -\frac{1}{\beta} \ln \left( \text{Tr}[\exp(-\beta\mathcal{H} + \sum_i \lambda_i \cdot (S_i - m_i))] \right). \quad (10)$$

Here,  $m_i$  is the average magnetization at site  $i$ ,  $m_i \equiv \langle S_i \rangle$  and  $\lambda_i$  is a Lagrange multiplier which fixes  $m_i$  to its mean-field value. The high-temperature expansion introduces fluctuations about the s-MFT solution. Defining  $\beta G(\beta) \equiv \tilde{G}(\beta)$ , the first two terms of the expansion in powers of  $\beta$ ,  $\tilde{G}(0)/\beta$  and  $\tilde{G}'(0)$ , are the entropy and energy at the s-MFT level, respectively. The prime represents differentiation with respect to  $\beta$ . The higher order terms in the  $\beta$  expansion of the Gibbs free-energy correspond to fluctuation corrections to the s-MFT free-energy that generate the terms that lift the  $U(1)$  degeneracy. We aim to calculate the higher order terms (beyond  $\beta^2$ ) in the  $\beta$  expansion of Eq. (10) that contribute to the degeneracy lifting term  $2\bar{\eta}_6|m_{xy}|^6 \cos(6\phi)$  of Eq. (8) to lowest order in the coupling constants  $j_{\pm\pm}$  and  $j_{z\pm}$ .<sup>57</sup> In other words, we are considering the selection near mean-field criticality and perturbative (in  $j_{z\pm}$  and  $j_{\pm\pm}$ ) vicinity of the  $U(1)$  symmetric portion of the theory ( $H_0$  in Eq. (1a)). Since we are focusing on the expansion of  $G$  in Eq. (10), we write its correction  $\delta G$  beyond the s-MFT solution as suggested by Eq.(8):

$$\delta G = 2\bar{\eta}_6(j_{\pm\pm}, j_{z\pm})|m_{xy}|^6 \cos(6\phi). \quad (11)$$

As discussed in Section II A,  $\bar{\eta}_6(j_{\pm\pm}, j_{z\pm}) = 0$  determines the phase boundaries between the  $\psi_2$  and the  $\psi_3$  states in the space of coupling constants  $j_{\pm\pm}$  and  $j_{z\pm}$ . We now proceed to calculate  $\bar{\eta}_6(j_{\pm\pm}, j_{z\pm})$  with the E-TAP method.

In the  $n^{\text{th}}$  order of the  $\beta$  expansion, factors of the form  $j_{\pm\pm}^k j_{z\pm}^l$  arise where  $k$  and  $l$  are positive integers and  $k+l = n$ . Each power of  $j_{\pm\pm}$  and  $j_{z\pm}$  contributes factors of  $e^{\pm 2i\phi}$  and  $e^{\pm i\phi}$ , respectively (see Eq. (1c)), to the corresponding term in the  $\beta$  expansion. By a simple power counting of these factors, one can pinpoint the terms that contribute to  $\cos(6\phi)$  and  $\cos(3\phi)$  that are necessary to compute due to their contribution to  $\omega$  and  $\eta_6$  and, consequently, to  $\bar{\eta}_6$  (see. Eq. (9)), and which arise at different orders in  $\beta$  in the E-TAP calculation. It is straightforward arithmetic to find what combinations of  $k$  and  $l$  in  $j_{\pm\pm}^k j_{z\pm}^l$  generate  $\cos(6\phi)$  and  $\cos(3\phi)$  terms. We then calculate  $\omega$  and  $\eta_6$  in Eqs. (5, 6) in terms of the microscopic couplings  $j_{\pm\pm}$  and  $j_{z\pm}$  using the E-TAP method (see Appendix C). We obtain  $\omega(j_{\pm\pm}, j_{z\pm})$ ,  $\eta_6(j_{\pm\pm}, j_{z\pm})$ , and thus  $\bar{\eta}_6(j_{\pm\pm}, j_{z\pm})$ , all to lowest nontrivial order in  $j_{\pm\pm}$  and  $j_{z\pm}$ . These read as:<sup>58</sup>

$$\omega(j_{\pm\pm}, j_{z\pm}) = a_0 \beta j_{\pm\pm} j_{z\pm} + \beta^2 (a_1 j_{\pm\pm}^2 j_{z\pm} + a_2 j_{z\pm}^3) + \beta^3 (a_3 j_{\pm\pm}^3 j_{z\pm} + a_4 j_{\pm\pm} j_{z\pm}^3) \quad (12)$$

and

$$\eta_6(j_{\pm\pm}, j_{z\pm}) = b_0 \beta^2 j_{\pm\pm}^3 + b_1 \beta^3 j_{\pm\pm}^2 j_{z\pm}^2 + b_2 \beta^4 j_{\pm\pm} j_{z\pm}^4 + b_3 \beta^5 j_{z\pm}^6, \quad (13)$$

and, consequently,

$$2\bar{\eta}_6(j_{\pm\pm}, j_{z\pm}) = c_0\beta^2 j_{\pm\pm}^3 + c_1\beta^3 j_{\pm\pm}^2 j_{z\pm}^2 + c_2\beta^4 j_{\pm\pm} j_{z\pm}^4 + \beta^5 (c_3 j_{\pm\pm}^4 j_{z\pm}^2 + c_4 j_{\pm\pm}^2 j_{z\pm}^4 + c_5 j_{z\pm}^6). \quad (14)$$

In Eqs. (12, 13, 14),  $a_i$  ( $i = 0, \dots, 4$ ),  $b_j$  ( $j = 0, \dots, 3$ ) and  $c_k$  ( $k = 0, \dots, 5$ ) are numerical coefficients determined by the explicit E-TAP calculation too lengthy to reproduce here, but whose derivation is described in Appendix C. Considering the highest power of coupling constants  $j_{\pm\pm}$  and  $j_{z\pm}$  in Eq. (14), one needs to compute terms up to sixth order in the high-temperature expansion of  $G$  in Eq. (10).

## 2. Results

In order to obtain the numerical value of the  $a_i$ ,  $b_j$  and  $c_k$  coefficients in Eq. (14), we employ a diagrammatic technique to represent the terms in the  $\beta$  expansion and which constitutes the computational core of the E-TAP method. These diagrams are composed of vertices and bonds.<sup>59</sup> The vertices correspond to lattice sites covered by a diagram and the bond represents the interaction between the vertices that it connects. The details necessary to carry out the calculations using diagrams are presented in Appendix C4. As mentioned earlier, for computational simplicity, we only consider the case of  $j_{zz} = 0$ . Recalling Eq. (9), and noting that to the lowest order we can use the s-MFT value of  $r_1$ ,  $r_1 = 2/\beta$ , we find:

$$2\bar{\eta}_6(j_{\pm\pm}, j_{z\pm}) = 10^{-5} \times [-6.50\beta^2 j_{\pm\pm}^3 + 57.3\beta^3 j_{\pm\pm}^2 j_{z\pm}^2 + 26.5\beta^4 j_{\pm\pm} j_{z\pm}^4 + 27.8\beta^5 j_{z\pm}^6 - 7.95\beta^5 j_{\pm\pm}^4 j_{z\pm}^2 - 4.60\beta^4 j_{\pm\pm}^3 j_{z\pm}^2 + 66.5\beta^5 j_{\pm\pm}^2 j_{z\pm}^4]. \quad (15)$$

$a_0$	$a_1$	$a_2$	$a_3$	$a_4$
-0.593	-1.13	-0.451	-0.938	7.21

TABLE I. Values of  $a_i$  in  $\omega(j_{\pm\pm}, j_{z\pm})$  multiplied by  $10^2$ .

$b_0$	$b_1$	$b_2$	$b_3$
-0.322	2.92	1.42	1.42

TABLE II. Values of  $b_j$  in  $\eta_6(j_{\pm\pm}, j_{z\pm})$  multiplied by  $10^4$ .

In deriving Eq. (15), we used  $\omega$  and  $\eta_6$  in Eqs. (12, 13), respectively, with the numerical values of  $a_i$  and  $b_j$  coefficients given in Tables I and II. Since  $\bar{\eta}_6(j_{\pm\pm}, j_{z\pm}) = 0$  determines the boundaries between the  $\psi_2$  and  $\psi_3$  states, we need to compute the roots of Eq. (15). In order to be consistent with the above E-TAP derivation perturbative in  $j_{z\pm}$  and  $j_{\pm\pm}$ , we ought to only consider the lowest order term in the root of  $\eta_6$  given by Eq. (15). We find that Eq. (15) has one real root which to, the lowest order in the coupling constants, reads:

$$j_{\pm\pm} \approx p_0 j_{z\pm}^2 + \dots \quad (16)$$

where the  $\dots$  represent higher order terms in  $j_{z\pm}$  and  $p_0 \approx 9.37\beta$  which determines the phase boundary between the  $\psi_2$  and the  $\psi_3$  states for  $j_{zz} = 0$ ,  $j_{z\pm} \ll 1$  and  $\beta \gtrsim \beta_c$ . The corresponding  $\psi_2$ - $\psi_3$  boundary for  $\beta = 0.253 \gtrsim \beta_c^{\text{MF}} = 1/4$ , providing an estimate of the boundary in the limit  $T \rightarrow T_c^-$ , is shown Fig. 2 The “outer” s-MFT boundaries of the overall

$\Gamma_5$  region for  $j_{zz} = 0$  is also shown in this figure. Outside the  $\Gamma_5$  boundary, the system orders in a Palmer-Chalker (PC) state<sup>17,26,60</sup> or a splayed ferromagnet (SF).<sup>45</sup>

It was found in Ref. [17] that, at zero-temperature, quantum fluctuations yield three distinct phase boundaries for  $j_{zz} = 0$ , a result confirmed in Ref. [32], even in the regime  $j_{z\pm} \ll 1$  and  $j_{\pm\pm} \ll 1$ , for which the E-TAP results for the ordered state selection at  $T \lesssim T_c$  presented here apply. The combination of the E-TAP results with those from Ref. [17] suggests, because of the different  $\psi_2/\psi_3$  boundaries at  $T = T_c^-$  and  $T = 0$ , the possibility of multiple transitions between  $\psi_2$  and  $\psi_3$  as the temperature is decreased well below  $T_c$  as was found for the model of Ref. [34]. Such a multiple-transition scenario constitutes an exotic variant of the more conventional ObD phenomenon since fluctuations select distinct long-range ordered states in different temperature regimes ( $T \lesssim T_c$  and  $T = 0^+$ ). That being said, one should be reminded that since the E-TAP phase diagram of Fig. 2 was constructed on the basis of a *lowest order* fluctuation correction to s-MFT described by Eqs. (12,13,14) and for a classical version of  $\mathcal{H}$  in Eq. (1), this discussion of multiple transitions is therefore only qualitative within the present E-TAP calculation. However, we expand further on this topic in Section IV within a Ginzburg-Landau theory framework. In the low-temperature regime,  $T \ll T_c$ , a study that incorporates high-order magnon-magnon interaction at nonzero temperature starting from the results of Ref. [17] would be of interest to explore this phenomenology further. This is, however, beyond the scope of the present work.

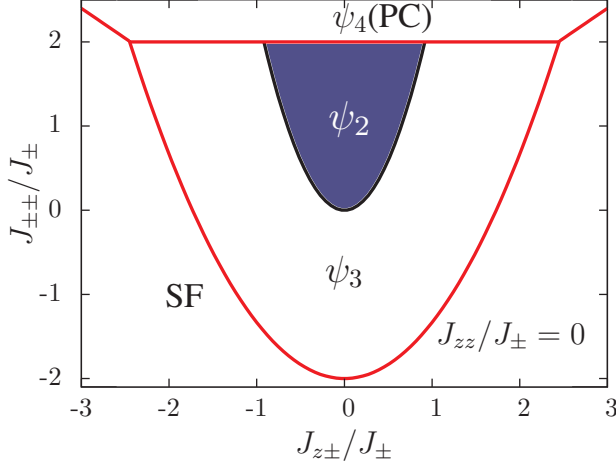


FIG. 2. (Color Online) The  $j_{zz} = 0$  phase diagram of the model at  $T \lesssim T_c$ . The  $\Gamma_5$  region corresponds to the region enclosing the  $\psi_2$  and  $\psi_3$  states taken together. The  $\Gamma_5$  region is circumscribed by an outer (red) parabola  $j_{\pm\pm} = 2j_{zz}^2/3 - 2$  and a horizontal red line at  $j_{\pm\pm} = 2$  obtained from the s-MFT calculation.<sup>17</sup> The phase boundary between the  $\psi_2/\psi_3$  states obtained from the E-TAP calculations (i.e. the roots of  $\tilde{\eta}_6 = 0$ , see text) is represented by the black line. The phases outside of the boundaries of the  $\Gamma_5$  region are: a splayed ferromagnet (SF)<sup>45</sup> canted from the  $[100]$  cubic direction and the so-called  $\psi_4$  or Palmer-Chalker (PC) state.<sup>17,26,60</sup> Both the SF and the PC phases have  $\mathbf{k} = 0$  ordering wave vector.

### B. Cluster mean-field theory (c-MFT)

It is interesting to ask to what extent a calculation, any calculation, that goes beyond s-MFT may reveal an ordered state selection at  $T \lesssim T_c$ . For this reason, we study in this section the effect of fluctuations numerically using c-MFT. This method incorporates fluctuations at the level of one tetrahedron by exactly diagonalizing the Hamiltonian corresponding to the tetrahedron and treating the interactions between different tetrahedra at the s-MFT level. The c-MFT approach does not involve a perturbative scheme in powers of  $\beta$  compared with the E-TAP method where fluctuations are included through an expansion of the Gibbs free-energy in powers of  $\beta$  (see Eq. (10)). In c-MFT, fluctuations are limited to one tetrahedron while, in the E-TAP method, fluctuations beyond one tetrahedron are included up to the range spanned by the diagrams generating the sixth order terms in the high-temperature expansion of  $G$  (see Appendix C 4 for details). One practical advantage of the c-MFT method compared to the E-TAP approach is that c-MFT allows us to easily investigate the effect of nonzero  $J_{zz}$  on the selection of the  $\psi_2$  versus the  $\psi_3$  state. In what follows we first provide the details of the c-MFT method and then present in Fig. 4 the results of the calculations for various  $J_{zz}$  values.

### I. Method

The c-MFT that we use here may be viewed as a general, non-perturbative and rather system-independent way to obtain (numerical) results beyond s-MFT. While the approach is not specific to the type of spins (classical or quantum) considered, we restrict ourselves to quantum spins 1/2 for a tetrahedron cluster. A pragmatic reason for using quantum spins here is to avoid the complicated eight integrals over the solid angle that correspond to the classical trace for classical spins taken as vectors of fixed length  $|\mathbf{S}_i| = 1/2$  and orientation defined by an azimuthal and a polar angle. To illustrate the method, we write the model Hamiltonian (1) in the compact form:

$$\mathcal{H} = \sum_{\langle ij \rangle, \mu, \nu} J_{ij}^{\mu\nu} S_i^\mu S_j^\nu, \quad (17)$$

with  $\mu, \nu = z, +, -$  and:

$$J_{ij}^{\mu\nu} := \begin{pmatrix} J_{zz} & J_{z\pm}\zeta_{ij} & J_{z\pm}\zeta_{ij}^* \\ J_{z\pm}\zeta_{ij} & J_{\pm\pm}\gamma_{ij} & -J_{\pm} \\ J_{z\pm}\zeta_{ij}^* & -J_{\pm} & J_{\pm\pm}\gamma_{ij}^* \end{pmatrix}. \quad (18)$$

For  $N$  sites on the pyrochlore lattice, there are  $N/4$  "up" and  $N/4$  "down" corner-sharing tetrahedra (see Fig. 3). The diamond lattice, dual to the pyrochlore lattice, offers a simple representation of the tetrahedra that tessellate the lattice as elementary units. We note that the diamond lattice is bipartite (it is composed of 2 FCC sublattices, say A and B). As we consider ordered phases with a  $\mathbf{k} = 0$  ordering wave vector, we assume in the c-MFT method that all tetrahedra on sublattice A are equivalent and interact with each other only through the mean fields. We re-express model (17) in a more suggestive form as a sum over sublattice A (labelled with  $I$  or  $I'$ ) and take  $i, j = 1, 2, 3, 4$  as the sublattice indices:

$$\mathcal{H} = \sum_{I \in A} \sum_{\langle ij \rangle, \mu, \nu} J_{ij}^{\mu\nu} S_{i,I}^\mu S_{j,I}^\nu + \sum_{\langle I, I' \rangle \in A} \sum_{\langle ij \rangle, \mu, \nu} J_{ij}^{\mu\nu} S_{i,I}^\mu S_{j,I'}^\nu \quad (19)$$

We then proceed to apply a s-MFT approximation *only* on the second term of the Eq.(19): we neglect any fluctuations *between* the A tetrahedra while taking full account of fluctuations *within* the A tetrahedra. The self-consistently determined mean fields  $\langle S_{i,I}^\mu \rangle \equiv m_{i,I}$  are introduced to decouple the inter-cluster bonds. Again, because we focus on the  $\Gamma_5$  manifold with  $\mathbf{k} = 0$  ordering wave vector, we have translational invariance of sublattice A which implies  $m_{i,I'}^\nu = m_{i,I}^\nu$ . By performing this approximation, Eq. (19) reduces to a sum over the A tetrahedra coupled together by the mean-fields:

$$\mathcal{H}_{\text{MF}} = \sum_{I \in A} \sum_{i,j,\mu,\nu} J_{ij}^{\mu\nu} \left( \frac{1}{2} S_{i,I}^\mu S_{j,I}^\nu + S_{i,I}^\mu m_{j,I}^\nu - \frac{1}{2} m_{i,I}^\mu m_{j,I}^\nu \right). \quad (20)$$

Any thermodynamic average is readily computed from  $\mathcal{H}_{\text{MF}}$ . In particular, it is straightforward to show that the physical



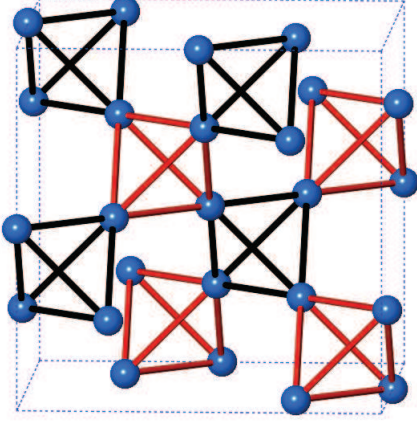


FIG. 3. The pyrochlore lattice, with spins located at the vertices of corner-sharing tetrahedra. There are two orientations of tetrahedra indicated by the black and red colored tetrahedra with the interaction paths represented by the colored lines. The centers of the tetrahedra form a diamond lattice (with sublattice A and B corresponding respectively to, say, the black and red tetrahedra). In c-MFT, the interaction in-between the black tetrahedra is treated at the mean-field theory level.

solution  $\{m_i^\mu\}$  that minimizes the free-energy corresponds to  $m_i^\mu = \langle S_i^\mu \rangle$ , where the thermodynamic average is taken with respect to the cluster mean-field Hamiltonian  $\mathcal{H}_{\text{MF}}$ . While we focus on the  $|S_i| = 1/2$  case, we note that c-MFT could be used for  $|S_i| > 1/2$  and extended to clusters with more than 4 spins. The method (using quantum spins) is only constrained by the computational limitations of the required exact full diagonalization over the cluster considered.<sup>61</sup>

## 2. Results

The critical temperature  $T_c$  is a function of the  $J_\pm$ ,  $J_{z\pm}$  and  $J_{\pm\pm}$  exchange parameters. For every point in the phase diagrams (i.e. for a given set of exchange couplings  $j_{z\pm}$  and  $j_{\pm\pm}$ ), we determine  $T_c$  by identifying the temperature at which there is a minimum of the free-energy and the development of a numerically non-zero on-site magnetization  $\mathbf{m}$ . For definitiveness, we then take a temperature slightly below  $T_c$ ,  $T = 0.9T_c$ , for each  $(j_{z\pm}, j_{\pm\pm})$  and determine which state ( $\psi_2$  or  $\psi_3$ ) is selected. We record this state selection over a grid of points that span the  $\Gamma_5$  manifold.

The c-MFT  $\psi_2/\psi_3$  phase boundary obtained for various  $j_{zz}$  values is presented in Fig 4. The previous symmetry analysis in Section II A of the  $\Gamma_5$  manifold showed that a nonzero  $m_z$  component is only compatible with the  $\psi_2$  state. As a corollary and confirmation of this expectation, we observe that as  $j_{zz}$  is varied, the  $\psi_2/\psi_3$  phase boundary position shifts. This can be understood physically in the following way. A negative  $j_{zz}$  favors a “4-in/4-out” hence non-zero  $m_z$  and thus the  $\psi_2$  state. Conversely, a positive  $j_{zz}$  on a single tetrahedron alone would favor a “2-in/2-out” spin configuration and therefore disfavor a “4-in/4-out” configuration. However, an

ordered state built with such “2-in/2-out” spin configuration would have a nonzero net magnetization on each tetrahedron, as well as for the whole system since we consider a  $\mathbf{k} = 0$  propagation vector, which is forbidden for the  $\Gamma_5$  representation under discussion. Therefore, the best that a positive  $j_{zz}$  can do is to favor the  $\psi_3$  state ( $m_z = 0$ ) against  $\psi_2$  ( $m_z \neq 0$ ) and the boundary shifts accordingly, as shown in Fig. 4.

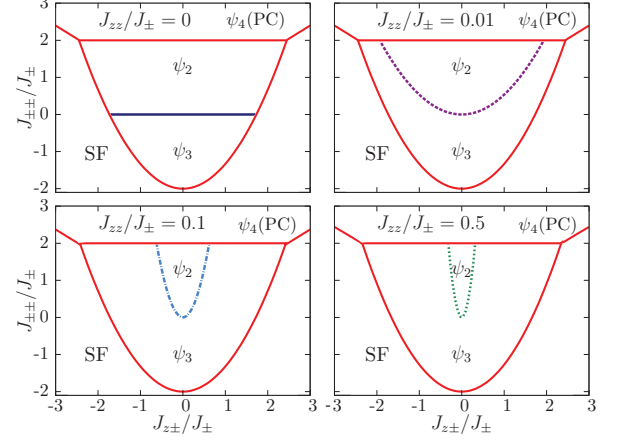


FIG. 4. (Color Online) The c-MFT phase diagram at  $T \lesssim T_c$  obtained for positive  $j_{zz} = 0.5, 0.1, 0.01, 0$ . The region circumscribed by the red line is the  $\psi_2/\psi_3$   $\Gamma_5$  region. For a given phase boundary (fixed  $j_{zz}$ ) the area above (below) that phase boundary line corresponds to the  $\psi_2$  ( $\psi_3$ ) phase. The phase boundaries for negative values of  $j_{zz}$  will be approximately a reflection of the presented phase boundaries with respect to the  $j_{zz} = 0$  line. Again, the phases outside of the boundaries of the  $\Gamma_5$  region are: a splayed ferromagnet (SF) canted from the [100] cubic direction<sup>45</sup> and the so-called  $\psi_4$ <sup>26</sup> or Palmer-Chalker (PC) state.<sup>60</sup>

A comment is warranted regarding the flat ( $J_{z\pm}$ -independent)  $\psi_2/\psi_3$  phase boundary predicted by c-MFT for  $J_{zz} = 0$  in Fig. 4. This is to be contrasted with the E-TAP results of Fig. 2 and the  $T = 0$  quantum fluctuation calculations of Ref. [17] (see Fig. 3 therein), which both show a  $J_{z\pm}$ -dependent  $\psi_2$ - $\psi_3$  phase boundary for  $J_{zz} = 0$ . This  $J_{z\pm}$  independent c-MFT result can be rationalized in the following way with the argument proceeding through a sequence of steps. In the c-MFT approximation that considers a one-tetrahedron cluster, the  $m_z$  dependence of the free-energy comes only from the  $J_{zz}$  term because of the special role the  $C_3$  symmetry plays when considering such a cluster. The reason is that every spin on a one-tetrahedron cluster is coupled to three identical “exterior” mean fields  $\mathbf{m}_{j,I}$  because of the  $\mathbf{k} = 0$   $\psi_2$  or  $\psi_3$  long-range ordered states considered. As a result, terms such as  $(\sum_i S_i^\pm)(\sum_j \zeta_{ij} m_{j,I}^\pm)$  vanish due to the  $C_3$  symmetry that imposes the necessary form for the  $\zeta_{ij}$  bond factors.<sup>16,45,49</sup> The same argument of course applies for terms of the form  $(\sum_i m_{i,I}^\pm)(\sum_j \zeta_{ij} S_j^\pm)$ . Consequently, the free-energy depends on  $m^z$  explicitly only via the  $J_{zz}$  coupling since the combined dependence on  $J_{z\pm}$  and  $m^z$  is eliminated via the above argument. When  $J_{zz} = 0$ , all dependence of the c-MFT free-energy on  $m^z$  disappears, and thus no-dependence on  $J_{z\pm}$  can remain, as found in Fig. 4. On



the other hand, when  $J_{zz} \neq 0$ , the c-MFT free-energy depends on  $m^z$ . In that case,  $J_{z\pm}$  coupling the  $S_i^z$  and  $S_i^\pm$  components will, through the *intra-tetrahedra* fluctuations that are incorporated into the c-MFT calculation, renormalize the effective intra-tetrahedron  $zz$  component sublattice susceptibility and thus the net effect of *inter-tetrahedra* mean-field ( $\sum_j J_{zz} m_{j,J}^z$ ). As a result, for  $J_{zz} \neq 0$ , a  $J_{z\pm}$  dependence of the  $\psi_2/\psi_3$  phase boundary is observed for a single-tetrahedron c-MFT calculation (see Fig. 4).

While the above argument for the  $J_{z\pm}$  independence of the boundary for the  $J_{zz} = 0$  case applies for a single tetrahedron cluster c-MFT, we expect the boundary for larger clusters to depend on  $J_{z\pm}$ , even for  $J_{zz} = 0$ . In that case,  $J_{z\pm}$  may induce a  $m^z$  dependence that would result in a behavior similar to E-TAP results for  $J_{zz} = 0$ . The reason is that, for larger clusters, the sites on the perimeter of the cluster may be coupled to the mean-field parameters  $m_{j,I}$  coming from less than three sites related by  $C_3$  symmetry, and which caused the inter-tetrahedron mean-field to vanish for a 4-site tetrahedron cluster as discussed above. Notwithstanding this caveat associated with the special symmetry of the 4-site tetrahedron cluster, we nevertheless believe that the evolution of the boundary upon varying  $J_{zz}$  shown in Fig. 4 to be roughly qualitatively correct for  $J_{zz} \neq 0$ .

#### IV. MULTIPLE TRANSITIONS AT $T < T_c$

The results above, along with those of Refs. [17 and 32], suggest that in some circumstances the state selected at  $T_c$  may differ from the low-temperature phase selected by harmonic (classical or quantum) spin waves. For example, Ref. [34] found that, in a classical pyrochlore Heisenberg antiferromagnet with additional indirect Dzyaloshinskii-Moriya interaction, the state selected at  $T \lesssim T_c$  is  $\psi_2$  while the one selected at  $T = 0^+$  is  $\psi_3$ . One might then ask what are the generic possibilities that may occur in  $xy$  pyrochlore magnets for which the anisotropic exchange terms position them in the degenerate  $\Gamma_5$  manifold. Considering the lowest order term in  $m_{xy}$ , one has, as discussed above in Section II A

$$\delta\mathcal{F}_{\text{GL}}(\phi) = f_6(T) \cos(6\phi). \quad (21)$$

$f_6(T) \equiv 2\bar{\eta}_6|m_{xy}|^6$  is a function of temperature,  $T$ , for fixed anisotropic exchange terms  $J_\pm$ ,  $J_{zz}$ ,  $J_{z\pm}$  and  $J_{\pm\pm}$ , with the sign of  $f_6(T)$  dictating which state is selected at a given temperature. If  $f_6(T = 0^+)$  stays of the same sign for all  $0 < T < T_c$ , only a single phase is realized below  $T_c$ . With solely this lowest order anisotropic term in  $\delta\mathcal{F}_{\text{GL}}$ , the only other possibility is a first order transition between  $\psi_2$  and  $\psi_3$  at some temperature  $T^* < T_c$  when  $f_6(T)$  changes sign at  $T = T^*$ . However, as one gets far below  $T_c$ , higher order terms in  $|m_{xy}|$  can become of significant magnitude in  $\mathcal{F}_{\text{GL}}$ . One may then extend the Ginzburg-Landau free-energy by incorporating higher-order harmonics in the anisotropy potential, as

$$\delta\mathcal{F}_{\text{GL}}(\phi) = \sum_{n=1} 2\bar{\eta}_{6n}(T)|m_{xy}|^{6n} \cos(6n\phi). \quad (22)$$

Keeping only the two lowest order terms for illustration purposes, we have

$$\delta\mathcal{F}_{\text{GL}}(\phi) = f_6(T) \cos(6\phi) + f_{12}(T) \cos(12\phi), \quad (23)$$

where  $f_{6n} \equiv 2\bar{\eta}_{6n}|m_{xy}|^{6n}$ . For simplicity, consider first  $f_{12} = -1$ . As  $f_6(T)$  varies from  $f_6(T) < 0$  to  $f_6(T) > 0$ , a first order transition occurs when the minimum of  $\delta\mathcal{F}_{\text{GL}}$  shifts discontinuously, in a first order transition, from  $\phi = 0$  ( $\psi_2$  state) to  $\phi = \pi/6$  ( $\psi_3$  state) (see Fig. 5a). A more interesting behavior is in principle possible. Consider now  $f_{12} = +1$ . In that case, second order (Ising-type) transitions occur at  $f_6 = \pm 4|f_{12}|$ . For example, for  $f_6 < -4$ , the minimum is at  $\phi = 0$  (a  $\psi_2$  state), and a second order transition to a  $f_6$ -dependent angle  $\phi$  occurs at  $f_6 = -4$ . As  $f_6$  continues growing and become less negative (at fixed  $f_{12} = +1$ ), the magnetic moment orientation  $\phi$  continues increasing until another second order transition to the  $\psi_3$  state occurs at  $f_6 = +4$  (see Fig. 5b). While the direct  $\psi_2$  to  $\psi_3$  transition observed in the pyrochlore Heisenberg antiferromagnet with indirect Dzyaloshinskii-Moriya interaction<sup>34</sup> suggests that this system belongs to the first case, the second scenario is perhaps not excluded in more complex models in which both temperature and applied magnetic field are varied simultaneously, or when the magnetic  $\text{Er}^{3+}$  sites are diluted by non-magnetic ions.<sup>62,63</sup>

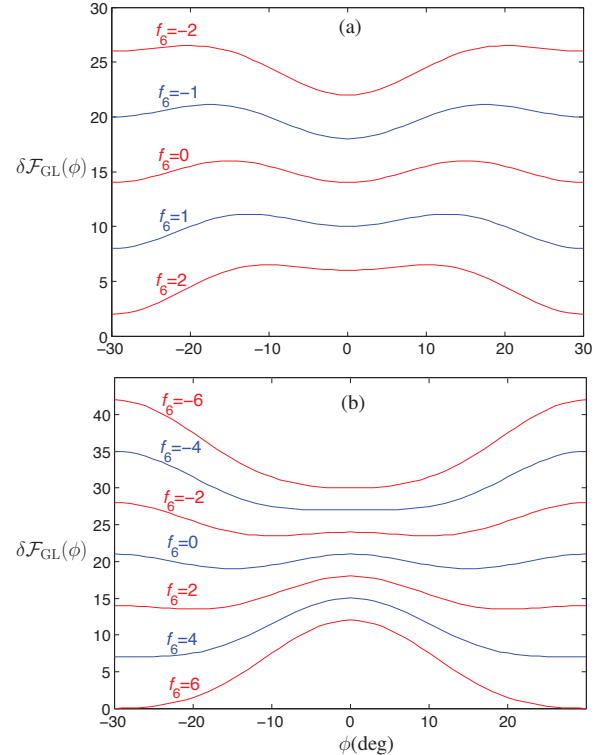


FIG. 5. (Color Online) (a) Sequence of  $\delta\mathcal{F}_{\text{GL}}(\phi)$  with  $f_{12} = -1$  and  $f_6 = -2, -1, 0, 1$  and  $2$  (from top to bottom). There is a first-order transition at  $f_6 = 0$ . (b) Sequence of  $\delta\mathcal{F}_{\text{GL}}(\phi)$  with  $f_{12} = 1$  and  $f_6 = -6, -4, -2, 0, 2, 4$  and  $6$  (from top to bottom). In mean-field theory, there are Ising-type second-order transitions at  $f_6 = \pm 4|f_{12}|$ .

In the above discussion, we have implicitly assumed that the anisotropic terms  $[m_z(m_{xy})^3 + \text{c.c.}]^k$  and  $[(m_{xy})^6 + \text{c.c.}]^l$ , which give terms of the form  $|m_{xy}|^{6n} \cos(6n\phi)$  in the Ginzburg-Landau theory once  $m_z$  has been eliminated (see discussion in Section II A), are generated by thermal fluctuations beyond the Ginzburg-Landau free-energy derived by a s-MFT treatment of the microscopic model  $\mathcal{H}$  in Eq. (1). However, it is in principle possible that virtual crystal field excitations (VCFE)<sup>31,44,46,47</sup> mediated by the bare multipolar interactions between the rare-earth ions would generate<sup>31,64</sup> an effective pseudospin-1/2 Hamiltonian more complex than the one given by Eqs. (1a), (1b) and (1c) and involve multispin (e.g. ring-exchange like) interactions capable of lifting degeneracy at the classical level without invoking order-by-disorder.<sup>15,16</sup> However, one naively expects those interactions generated by VCFE to be significantly smaller compared with the  $J_\pm$ ,  $J_{\pm\pm}$ ,  $J_{z\pm}$  and  $J_{zz}$  of Eq. (1a), as stated in Ref. 16. Whether those terms are truly inefficient in competing with order-by-disorder, once the commonly large prefactors of combinatoric origin arising in high-order perturbation theory have been taken into account, must await a detailed calculation.

## V. DISCUSSION

In this work, we first used an extended TAP (E-TAP) method to analytically study the problem of order-by-disorder (ObD) near the critical temperature of a general three-dimensional  $xy$  antiferromagnetic model on the pyrochlore lattice with the Hamiltonian of Eq. (1) with  $J_{zz} = 0$ . We focused on the  $\Gamma_5$  manifold which is  $U(1)$  degenerate at the standard mean-field theory (s-MFT) level. The fluctuations corrections to the free-energy beyond s-MFT were organized as an expansion in powers of the inverse temperature,  $\beta$ , and to lowest orders in the coupling constants  $j_{\pm\pm} = J_{\pm\pm}/J_\pm$ ,  $j_{z\pm} = J_{z\pm}/J_\pm$  and the on-site magnetization  $m$ . We established that in different parts of the  $\Gamma_5$  region, the  $\psi_2$  and  $\psi_3$  states are the only minima of the free-energy selected by fluctuations up to the lowest order  $U(1)$  symmetry-breaking terms considered. The phase boundary between  $\psi_2$  and  $\psi_3$  can then be obtained by finding the real roots of Eq. (14) in terms of  $J_{\pm\pm}/J_\pm$  and  $J_{z\pm}/J_\pm$ . We also numerically studied the ObD mechanism in the 3D- $xy$  pyrochlore system using cluster mean-field theory (c-MFT). Using this method, we obtained a phase boundary between  $\psi_2$  and  $\psi_3$  states for a variety of  $J_{zz}$  values shown in Fig. 4.

Using a Ginzburg-Landau theory, along with E-TAP and c-MFT calculations, we predict that for a state ordered in the local  $xy$  plane at the s-MFT level, fluctuations can induce a small out-of- $xy$ -plane  $m_z$  component of the on-site magnetization. This fluctuation-induced local  $z$  component of the magnetization is only compatible with the  $\psi_2$  state. We expect the size of this moment to be small, since it is proportional to  $|m_{xy}|^3 \ll 1$  for  $T \lesssim T_c^{\text{MF}}$  (see Eq. (7)). Yet, on the basis of the c-MFT results, we might anticipate that nonzero  $m_z$  has an important effect on the phase boundary between the  $\psi_2$  and the  $\psi_3$  states for  $J_{zz} \neq 0$ , as illustrated in Fig. 4 (see also the

caption of the figure).

By considering the single phase boundary between the  $\psi_2$  and  $\psi_3$  states for  $T \lesssim T_c$  along with the multiple  $\psi_2$ - $\psi_3$  boundaries found at  $T = 0^+$  in Ref. [17], for  $j_{\pm\pm} \ll 1$  and  $j_{z\pm} \ll 1$ , the regime for which the perturbative E-TAP solutions above apply, one may expect to find multiple phase transitions between  $\psi_2$  and  $\psi_3$  states upon decreasing temperature from  $T \lesssim T_c$  to  $T \sim 0^+$ . While possible in principle and found in a numerical study,<sup>65</sup> such multiple transitions in real  $xy$  pyrochlore magnetic materials have not yet been reported. Since in the E-TAP calculations the  $j_{\pm\pm}$  and  $j_{z\pm}$  couplings were treated perturbatively, the E-TAP phase boundary in Eq. (16) is valid for small  $j_{\pm\pm}$  and  $j_{z\pm}$  and would thus need to be modified for non-perturbative  $j_{\pm\pm}$  and  $j_{z\pm}$ , that is, closer to the boundaries of the  $\Gamma_5$  region with the splayed-ferromagnet (SF) phase (see Fig. (2)). A case in point is the Heisenberg pyrochlore antiferromagnet with indirect Dzyaloshinskii-Moriya interaction.<sup>65</sup> This model displays a transition at  $T_c^+$  from a paramagnetic state to  $\psi_2$ , followed at  $T_c^- < T_c^+$  by a transition from  $\psi_2$  to  $\psi_3$ ,<sup>65</sup> and for which the corresponding anisotropic exchange  $J_\pm$ ,  $J_{\pm\pm}$ ,  $J_{z\pm}$  and  $J_{zz}$  are such that this model lives *right on* the  $\Gamma_5$  to SF classical phase boundary.<sup>17</sup>

The difference between the phase boundaries obtained by E-TAP and c-MFT for  $J_{zz} = 0$  (see Figs. 2 and 4) arises from the different range of fluctuations considered in these two methods. In c-MFT, because of the exact diagonalization of the Hamiltonian on a single tetrahedron and the s-MFT treatment of the inter-tetrahedra couplings, the fluctuation corrections considered are short-ranged (limited to the nearest-neighbors). However in the E-TAP method, by considering the higher order terms in the  $\beta$  expansion, specifically  $\beta^4$  and beyond where larger (more extended) diagrams appear in the calculation (see Appendix C, Section C4), fluctuations beyond nearest-neighbors are included. It would be of interest to benchmark these arguments by performing c-MFT calculations for larger clusters.

The E-TAP corrections to the s-MFT free-energy could also be computed for the  $J_{zz} \neq 0$  case. The procedure for considering  $J_{zz} \neq 0$  is conceptually the same as for the  $J_{zz} = 0$  case for which, using the E-TAP method, we calculated the terms in the high temperature expansion of Eq. (10) that contribute to the degeneracy-lifting  $\cos(3\phi)$  and  $\cos(6\phi)$  terms in the free-energy. To obtain these terms to the lowest order in the coupling constants  $J_{\pm\pm}$ ,  $J_{z\pm}$  and  $J_{zz}$ , one would need to consider the high temperature expansion of  $G$  up to the seventh order in  $\beta$  and calculate the degeneracy lifting terms following the power-counting prescription explained above Eq. (12). However, the number of terms of the form  $j_{zz}^l j_{\pm\pm}^m j_{z\pm}^n$ , with fixed  $l + m + n$  value  $l$ ,  $m$  and  $n$  positive integers, and thus the number of diagrams to be computed, proliferate significantly as to make this calculation a significant undertaking. The  $J_{zz} \neq 0$  case thus stands on its own as an independent future study.

Finally, we remark that variants of the E-TAP method presented in this work could be applied to models other than the pyrochlore structure to analytically investigate the role of fluctuations beyond s-MFT in selecting a specific long-

range ordered state close to the critical temperature. One example is the case of Heisenberg spins on face centered cubic (FCC) lattice interacting via magnetostatic dipole-dipole interaction.<sup>66,67</sup> Generally speaking, one might expect that a consideration of an E-TAP analytical description of fluctuations at the microscopic level may help shed light on the role of individual symmetry-allowed interactions in the ordered state selection near the critical temperature over a broad range of frustrated spins models for which an accidental degeneracy exists in a standard mean-field theory description.

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## Appendix A: $\psi_2$ and $\psi_3$ states

The  $\psi_2$  states have spin configurations with a  $\mathbf{k} = 0$  ordering wave vector and with the following spin orientations on a tetrahedron expressed in the global (Cartesian) reference frame

$$\hat{e}_0^1 = \frac{1}{\sqrt{6}}(-1, -1, 2) \quad (\text{A1a})$$

$$\hat{e}_1^1 = \frac{1}{\sqrt{6}}(1, 1, 2) \quad (\text{A1b})$$

$$\hat{e}_2^1 = \frac{1}{\sqrt{6}}(1, -1, -2) \quad (\text{A1c})$$

$$\hat{e}_3^1 = \frac{1}{\sqrt{6}}(-1, 1, -2) \quad (\text{A1d})$$

Here the subscripts correspond to the four sublattice labels in the pyrochlore lattice and the superscripts refer to different symmetry-related  $\psi_2$  states.  $\{\hat{e}_i^2\}$  and  $\{\hat{e}_i^3\}$ ,  $i = 0, \dots, 3$ , can be obtained from Eq. (A1) by  $C_3$  ( $\pi/3$ ) rotations with respect to the  $\langle 111 \rangle$  directions.  $\psi_3$  states can be obtained from the  $\psi_2$  ones, by a  $\pi/6$  rotation of each spin about its local  $[111]$  axis.

## Appendix B: Symmetry groups of $\psi_2$ and $\psi_3$ states

The symmetries of either  $\psi_i$  state ( $i = 2, 3$ ) form a group known as the little group of the corresponding state. The generators of the  $\psi_3$  little group include the  $C_2$  rotation by  $\pi$  about one of the cubic  $x$ ,  $y$  or  $z$  axes (depending on the particular  $\psi_3$

state), the improper rotation by  $\pi/2$  ( $S_4$ ) about the same cubic axis, and two plane reflections  $\sigma_1$  and  $\sigma_2$  with respect to planes spanned by the cubic axis and one of the two tetrahedron bonds perpendicular to the cubic axis. The generators of the little group of  $\psi_2$  states include  $\tau S_4$ ,  $\tau \sigma_1$  and  $\tau \sigma_2$  where  $\tau$  is the time-reversal symmetry operation.

Considering the action of the symmetry operations in the  $\psi_2$ 's and  $\psi_3$ 's little groups on the possible configurations with a finite onsite  $m_z$  moments on a single tetrahedron (i.e. all-in/all-out, 2-in/2-out, 3-in/1-out and 1-in/3-out where “in” and “out” indicates whether  $m_{i,z}$  is positive or negative on site  $i$ ), only all-in-all-out configuration is invariant under the symmetry operations of the  $\psi_2$ 's little group while none of the above configurations are invariant under the  $\psi_3$ 's little group symmetry operations. As a result, only the  $\psi_2$  state can possess a finite onsite  $m_z$  moment which displays an all-in-all-out configuration on a single tetrahedron and therefore does not produce a net magnetic moment on a tetrahedron.

## Appendix C: Extended TAP Method (E-TAP)

In this Appendix, we first derive the general form of the E-TAP corrections up to sixth order in  $\beta$ . Next, we illustrate our method for calculating the terms in the  $\beta$  expansion of the Gibbs free-energy,  $G$ , of Eq. (10) by focusing on two calculation examples. Finally, we discuss the general properties of the diagrammatic approach employed to carry out the computation of the various terms entering the E-TAP calculation.

### 1. Derivation

The E-TAP method is based on a perturbative expansion of the Gibbs free-energy as a function of inverse temperature,  $\beta$ , beyond that given by s-MFT. There are several ways of deriving these corrections.<sup>68</sup> Here, we employ and extend the method presented by Georges and Yedidia<sup>41</sup> for Ising spin glass which provides the corrections due to fluctuations about the s-MFT solution, order by order in  $\beta$ . In this part of the Appendix, we focus on a temperature regime close to the mean-field transition temperature,  $T \lesssim T_c^{\text{MF}}$ , and derive the E-TAP corrections for classical Heisenberg spins of fixed length  $|\mathbf{S}| = 1/2$ .

The Hamiltonian in Eq. (1),  $\mathcal{H}$ , can be written as:

$$\mathcal{H} = \sum_{i < j} S_i^\mu J_{ij}^{\mu\nu} S_j^\nu, \quad (\text{C1})$$

Greek labels represent Cartesian coordinates and implicit summation over repeated superscripts is used. A Taylor series expansion of Eq. (10) in powers of  $\beta$  reads:

$$G(\beta) = \frac{1}{\beta} \left( \tilde{G}(\beta) \Big|_{\beta=0} + \frac{\partial \tilde{G}(\beta)}{\partial \beta} \Big|_{\beta=0} \beta + \frac{1}{2!} \frac{\partial^2 \tilde{G}(\beta)}{\partial \beta^2} \Big|_{\beta=0} \beta^2 \dots \right), \quad (\text{C2})$$

where  $\tilde{G}(\beta) = \beta G(\beta)$ . We define,

$$U \equiv \frac{1}{2} \sum_{ij} \delta S_i^\mu J_{ij}^{\mu\nu} \delta S_j^\nu \quad (\text{C3})$$

$$\frac{\partial(\beta G(\beta))}{\partial \beta} = \langle \mathcal{H} \rangle, \quad (\text{C5a})$$

$$\frac{\partial^2(\beta G(\beta))}{\partial \beta^2} = -\langle U^2 \rangle, \quad (\text{C5b})$$

$$\frac{\partial^3(\beta G(\beta))}{\partial \beta^3} = \langle U^3 \rangle, \quad (\text{C5c})$$

$$\frac{\partial^4(\beta G(\beta))}{\partial \beta^4} = -\langle U^4 \rangle + 3\langle U^2 \rangle^2 - 3\langle U^2 T_2 \rangle, \quad (\text{C5d})$$

$$\frac{\partial^5(\beta G(\beta))}{\partial \beta^5} = \langle U^5 \rangle - 10\langle U^2 \rangle \langle U^3 \rangle - 3\langle U^2 T_3 \rangle + 7\langle U^3 T_2 \rangle + 6\langle U T_2^2 \rangle, \quad (\text{C5e})$$

$$\begin{aligned} \frac{\partial^6(\beta G(\beta))}{\partial \beta^6} = & -\langle U^6 \rangle + 15\langle U^4 \rangle \langle U^2 \rangle + 10\langle U^3 \rangle^2 - 30\langle U^2 \rangle^3 - 12\langle U^4 T_2 \rangle \\ & + 10\langle U^3 T_3 \rangle - 3\langle U^2 T_4 \rangle - 27\langle U^2 T_2^2 \rangle + 18\langle U T_2 T_3 \rangle \\ & - 6\langle U T_3 \rangle \langle U^2 \rangle + 51\langle U^2 \rangle \langle U^2 T_2 \rangle + 6\langle U^2 \rangle \langle T_2^2 \rangle - 6\langle T_2^3 \rangle, \end{aligned} \quad (\text{C5f})$$

and where  $T_n$  is defined as

$$T_n \equiv \sum_i \frac{\partial^n \lambda_i}{\partial \beta^n} \cdot \delta S_i. \quad (\text{C6})$$

Since  $\partial \lambda / \partial \beta|_{\beta=0} = \mathbf{h}_i$ <sup>41</sup> where  $\mathbf{h}_i^\mu = \sum_{j,\nu} J_{ij}^{\mu\nu} m_j^\nu$ , the terms involving  $T_n$  come from considering fluctuations of the local mean-field. On the other hand, the  $U^n$  terms take into account the fluctuations of the on-site magnetization (see Eqs. (C3, C4)) within the ensemble set by the s-MFT solution. The  $\langle \dots \rangle$  above denotes a thermal average. For a general observable  $O$ ,  $\langle O \rangle$  is given by:

$$\langle O \rangle = \frac{\text{Tr}[O \exp(-\beta H + \sum_i \lambda_i \cdot (\mathbf{S}_i - \mathbf{m}_i))]}{\text{Tr}[\exp(-\beta H + \sum_i \lambda_i \cdot (\mathbf{S}_i - \mathbf{m}_i))]} \quad (\text{C7})$$

The first two terms in Eq. (C2) correspond to the s-MFT free-energy while the higher order terms in  $\beta$  provide corrections beyond s-MFT. Calculating the expectation value of powers of  $U$  at  $\beta = 0$  reduces to the evaluation of mean-field averages of the form:

$$\langle \delta S_{i_1}^{\alpha_1} \delta S_{i_2}^{\alpha_2} \dots \delta S_{i_n}^{\alpha_n} \rangle_{\text{MF}}, \quad (\text{C8})$$

where  $i_n$  represents the site label and  $\alpha_n$  represents a Cartesian coordinate.  $n$  is the number of  $\delta S$  factors in Eq. (C8). From now on, we drop the MF subscript for compactness of notation. For  $n = 1$ , the expectation value in Eq. (C8) is zero due to the relation  $\mathbf{m}_i = \langle \mathbf{S}_i \rangle$ . For  $n \geq 2$ , however, Eq. (C8)

where

$$\delta S_j^\nu \equiv S_j^\nu - m_j^\nu \quad (\text{C4})$$

are spin fluctuations about the s-MFT solution. As shown in Ref. [41], the derivatives of  $\tilde{G}(\beta)$  with respect to  $\beta$  can be evaluated in terms of expectation values of powers of  $U$  and  $T_n$ , that read:

is nonzero only if there is no site label that appears only once. For example, averages of the following form have a nonzero contribution:

$$\langle \delta S_i^{\alpha_1} \delta S_i^{\alpha_2} \delta S_j^{\alpha_3} \delta S_j^{\alpha_4} \delta S_j^{\alpha_5} \rangle = \langle \delta S_i^{\alpha_1} \delta S_i^{\alpha_2} \rangle \langle \delta S_j^{\alpha_3} \delta S_j^{\alpha_4} \delta S_j^{\alpha_5} \rangle \quad (\text{C9})$$

The expectation values above can be calculated using the self-consistent s-MFT equations for 3-component classical spins which are given by the Langevin function

$$\mathbf{m}_i = \frac{\lambda_i}{|\lambda_i|} \left[ \coth(|\lambda_i|) - \frac{1}{|\lambda_i|} \right]. \quad (\text{C10})$$

Consequently

$$\langle \delta S_i^\alpha \delta S_i^\beta \rangle = \frac{\partial m_i^\alpha}{\partial \lambda_i^\beta} \equiv \chi_i^{\alpha\beta}, \quad (\text{C11})$$

$$\langle \delta S_i^\alpha \delta S_i^\beta \delta S_i^\gamma \rangle = \frac{\partial \chi_i^{\alpha\beta}}{\partial \lambda_i^\gamma}, \quad (\text{C12})$$

and in general

$$\langle \delta S_i^{\alpha_1} \delta S_i^{\alpha_2} \dots \delta S_i^{\alpha_n} \rangle = \frac{\partial \langle \delta S_i^{\alpha_1} \delta S_i^{\alpha_2} \dots \delta S_i^{\alpha_{n-1}} \rangle}{\partial \lambda_i^{\alpha_n}}. \quad (\text{C13})$$

Since we are interested in a temperature range close to  $T_c$ , Eq. (C10) can be expanded for small  $|\lambda_i|$ :

$$m_i^\alpha = \frac{\lambda_i^\alpha}{3} \left[ 1 - \frac{(|\lambda_i|)^2}{15} + \frac{2(|\lambda_i|)^4}{315} + \dots \right], \quad (\text{C14})$$



The expansions of Eqs. (C11, C12, C13) at  $T \lesssim T_c$  can be computed by differentiating Eq. (C14) with respect to different components of the vector  $\lambda_i$  Lagrange multiplier.

In the rest of this section, we focus on the specific problem of the s-MFT  $U(1)$  degeneracy of the  $\Gamma_5$  manifold displayed by the Hamiltonian of Eq. (1) in a wide range of the  $\{J_\pm, J_{\pm\pm}, J_{z\pm}, J_{zz}\}$  anisotropic exchange parameters.<sup>17</sup> Below, we calculate the degeneracy-lifting contributions from  $\langle U^2 \rangle$  and  $\langle U^2 T_2 \rangle$  in Eqs. (C5b, C5d) to demonstrate the general idea of the method. Higher order terms can be calculated using a similar procedure.

## 2. $\langle U^2 \rangle$ Calculation

Using Eq. (C3), we have

$$\langle U^2 \rangle = \frac{1}{2^2} \sum_{\langle i_1, j_1 \rangle} \sum_{\langle i_2, j_2 \rangle} J_{i_1 j_1}^{\alpha_1 \beta_1} J_{i_2 j_2}^{\alpha_2 \beta_2} \langle \delta S_{i_1}^{\alpha_1} \delta S_{j_1}^{\beta_1} \delta S_{i_2}^{\alpha_2} \delta S_{j_2}^{\beta_2} \rangle, \quad (\text{C15})$$

where the summations are performed over lattice sites and summation is implied for Greek superscripts. To proceed, we need to specify all the possible pairings of the factors  $\delta S_{i_t}$ ,  $\delta S_{j_t}$  ( $t = 1, 2$ ) in the  $\langle \delta S_{i_1}^{\alpha_1} \delta S_{j_1}^{\beta_1} \delta S_{i_2}^{\alpha_2} \delta S_{j_2}^{\beta_2} \rangle$ .

Considering the description provided above Eq. (C9), the only nonzero pairings of site indices in Eq. (C15) are  $i_1 = i_2$ ,  $j_1 = j_2$  and  $i_1 = j_2$ ,  $j_1 = i_2$ , with the constraint  $i_1 \neq j_1$  and  $i_2 \neq j_2$  imposed by the Hamiltonian;  $J_{ii}^{\alpha\beta} = 0$  for all  $\alpha$  and  $\beta$ . This leads to:

$$\langle U^2 \rangle = \frac{1}{2} \sum_{\langle i, j \rangle} J_{ij}^{\alpha_1 \beta_1} J_{ij}^{\alpha_2 \beta_2} \langle \delta S_i^{\alpha_1} \delta S_i^{\alpha_2} \rangle \langle \delta S_j^{\beta_1} \delta S_j^{\beta_2} \rangle. \quad (\text{C16})$$

The computational complexity of the method requires the usage of a diagrammatic approach, which we now introduce by considering, for example, the calculation of Eq. (C16). In this equation, for a given  $i, j, \alpha_t$  and  $\beta_t$  with  $t = 1, 2$ ,  $J_{ij}^{\alpha_1 \beta_1} J_{ij}^{\alpha_2 \beta_2} \langle \delta S_i^{\alpha_1} \delta S_i^{\alpha_2} \rangle \langle \delta S_j^{\beta_1} \delta S_j^{\beta_2} \rangle$  can be represented by a diagram of the form illustrated in Fig. 6a. In this figure, the vertex labels match the labels of the lattice sites that the diagrams cover. Each vertex represents an average of the form  $\langle \delta S_i^{\alpha_1} \dots \delta S_i^{\alpha_t} \rangle$  where  $t$  is the number of bonds (solid lines) connected to that vertex. In the case of Eq. (C16) as indicated in Fig. 6a, the averages are  $\langle \delta S_i^{\alpha_1} \delta S_i^{\alpha_2} \rangle$  and  $\langle \delta S_j^{\beta_1} \delta S_j^{\beta_2} \rangle$ . The bonds represent the elements of the coupling matrix,  $J_{ij}^{\alpha\beta}$ . It is straightforward combinatorics to take into account only the terms that contribute to the degeneracy-lifting factors,  $\cos(3\phi)$  or  $\cos(6\phi)$ . We next proceed to demonstrate this point.

Based on the symmetry analysis of Section II A, we note that since  $\langle U^2 \rangle$  involves four powers of  $\delta S_i^{\alpha_t}$ ,  $\langle U^2 \rangle$  can only contribute degeneracy-lifting terms of the general form  $\propto m_z |m_{xy}|^3 \cos(3\phi)$  and where the proportionality factor is generated by the anisotropic couplings  $j_{\pm\pm}$  and  $j_{z\pm}$  in Eq. (1c). We recall that  $j_{\pm\pm}$  and  $j_{z\pm}$  each contributes a factor  $e^{\pm 2i\phi}$  and  $e^{\pm i\phi}$  in the power counting method. The  $-i\phi$  and  $-2i\phi$  corresponds to presence of  $\delta S^-$  and  $(\delta S^-)^2$  in Eq. (C16), respectively, which in turn, implies the presence of  $J_{ij}^{--} \equiv j_{\pm\pm} \gamma_{ij}^*$  or  $J_{ij}^{z-} \equiv j_{z\pm} \zeta_{ij}^*$  matrix elements

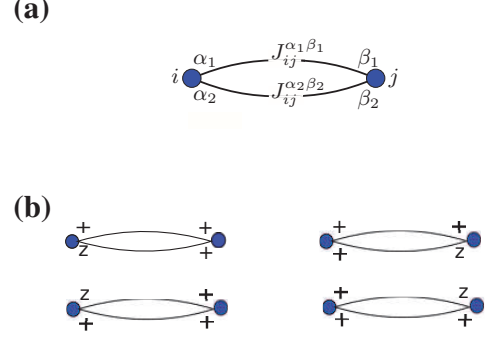


FIG. 6. **a)** Diagrammatic representation of the term  $J_{ij}^{\alpha_1 \beta_1} J_{ij}^{\alpha_2 \beta_2} \langle \delta S_i^{\alpha_1} \delta S_i^{\alpha_2} \rangle \langle \delta S_j^{\beta_1} \delta S_j^{\beta_2} \rangle$  in Eq. (C16). **b)** All diagrams corresponding to  $\alpha_i, \beta_i = z, +$ ,  $i = 1, 2$  with  $z$  appearing only once among the Greek superscripts. The same number of diagrams are present for the case where  $\alpha_i, \beta_i = z, -$ . Again  $z$  appears only once.

in  $J_{ij}^{\alpha_1 \beta_1} J_{ij}^{\alpha_2 \beta_2} \langle \delta S_i^{\alpha_1} \delta S_i^{\alpha_2} \rangle \langle \delta S_j^{\beta_1} \delta S_j^{\beta_2} \rangle$ . So for a given  $i$  and  $j$ , only the following combination of terms can generate  $\cos(3\phi)$ :

$$J_{ij}^{++} J_{ij}^{z+} \langle \delta S_i^+ \delta S_i^+ \rangle \langle \delta S_j^z \delta S_j^+ \rangle + J_{ij}^{--} J_{ij}^{z-} \langle \delta S_i^- \delta S_i^- \rangle \langle \delta S_j^z \delta S_j^- \rangle, \quad (\text{C17})$$

where the first term generates the  $e^{3i\phi}$  contribution to  $\cos(3\phi)$  while the second term generates  $e^{-3i\phi}$ . All possible ways of generating the factor  $e^{3i\phi}$  are illustrated in Fig. 6b, which is the same as the number of ways of generating  $e^{-3i\phi}$ . Ultimately, Eq. (C16) can be rewritten as:

$$\langle U^2 \rangle = 2j_{\pm\pm} j_{z\pm} \left( \langle \delta S^z \delta S^+ \rangle \langle \delta S^+ \delta S^+ \rangle \sum_{ij} \gamma_{ij} \zeta_{ij} + \text{h.c.} \right). \quad (\text{C18})$$

Based on Eqs. (C11, C14) and recalling from Section II that  $m$  and  $\phi$  do not require site indices in the  $\Gamma_5$  manifold, we have dropped the site labels of  $\delta S$ 's in Eq. (C16) when writing Eq. (C18). The lattice sum,  $\sum_{ij} \gamma_{ij} \zeta_{ij}$ , can be carried out using a computer program for different lattice sizes with linear dimension  $L$ . Up to sixth order in the  $\beta$  expansion, the lattice sums *per site* for different terms in Eqs. (C5) are independent of  $L$  for  $L \geq 2$ . Accordingly, we have

$$\frac{1}{N} \sum_{ij} \gamma_{ij} \zeta_{ij} = -6. \quad (\text{C19})$$

Using Eqs. (C11, C14), the averages in Eq. (C18) can be written as

$$\langle \delta S^z \delta S^+ \rangle = \frac{18}{45} m_z m_+ + \dots \quad (\text{C20})$$

$$\langle \delta S^+ \delta S^+ \rangle = \frac{18}{45} m_+^2 + \dots \quad (\text{C21})$$

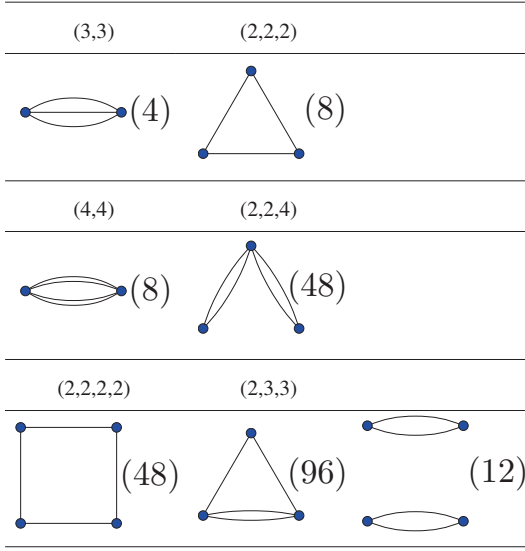


FIG. 7. For details, see the text in Appendix C 4. We also note that the contribution of disconnected diagrams (e.g. the diagram at the bottom right corner) in Eq. (C5) adds up to zero.

where to the lowest order of interest in  $\mathbf{m}$  which, in this case, is the fourth order, we kept  $\lambda^\alpha \simeq 3m^\alpha$  and neglect all higher order terms. Finally Eq. (C18) gives

$$\begin{aligned} \langle U^2 \rangle / N &= (-0.96m_z m_+^3 + \text{h.c.}) j_{\pm\pm} j_{z\pm} \\ &= -1.92 j_{\pm\pm} j_{z\pm} m_z |m_{xy}|^3 \cos(3\phi). \end{aligned} \quad (\text{C22})$$

We note that Eq. (C22) contributes to  $\omega$  in Eq. (6) which, in turn, is necessary to evaluate  $\bar{\eta}_6(j_{\pm\pm}, j_{z\pm})$  in Eq. (11). All other terms in Eq. (C5) involving solely powers of  $U$  and no  $T_n$  terms can be calculated in a similar way. The number of diagrams increases as one considers higher order terms in the  $\beta$  expansion of Eq. (10). As a result, finding the number and type of nonzero average of the form of Eq. (C8) is most easily done using a computer program. The details of this type of calculations are presented in Appendix C 4. Some of the diagrams that appear at higher order in the  $\beta$  expansion are illustrated in Fig. 7.

### 3. $\langle U^2 T_2 \rangle$ Calculation

Due to the presence of  $T_n$  in Eq. (C6), in this case  $n = 2$ , averages that involve  $T_n$  need to be carried out slightly differently in comparison with averages containing only powers of  $U$ . The difference comes from the presence of the factor  $\partial^n \lambda_i / \partial \beta^n$  in Eq. (C6). Considering Eq. (C5a) and the relation  $\partial \lambda / \partial \beta|_{\beta=0} = \mathbf{h}_i$ ,<sup>41</sup> we obtain the following relation

$$\frac{\partial \lambda^\alpha}{\partial \beta} = \frac{\partial \langle H \rangle}{\partial m^\alpha} = \frac{\partial^2 (\beta G(\beta))}{\partial m^\alpha \partial \beta}, \quad (\text{C23})$$

From Eq. (C23), one can write

$$\frac{\partial^n \lambda^\alpha}{\partial \beta^n} = \frac{\partial^{n+1} (\beta G(\beta))}{\partial m^\alpha \partial \beta^n}. \quad (\text{C24})$$

Using Eq. (C6),  $\langle U^2 T_2 \rangle$  can therefore be written as:

$$\langle U^2 T_2 \rangle = \frac{1}{2^2} \sum_{i_1 j_1} \sum_{i_2 j_2} \sum_k J_{i_1 j_1}^{\alpha_1 \beta_1} J_{i_2 j_2}^{\alpha_2 \beta_2} \frac{\partial^2 \lambda_k^{\alpha_3}}{\partial \beta^2} \langle \delta S_{i_1}^{\alpha_1} \delta S_{j_1}^{\beta_1} \delta S_{i_2}^{\alpha_2} \delta S_{j_2}^{\beta_2} \delta S_k^{\alpha_3} \rangle. \quad (\text{C25})$$

Using Eq. (C24), we have

$$\frac{\partial^2 \lambda_k^{\alpha_3}}{\partial \beta^2} = \frac{\partial^3 (\beta G(\beta))}{\partial m^{\alpha_3} \partial \beta^2} = -\frac{\partial \langle U^2 \rangle}{\partial m^{\alpha_3}}. \quad (\text{C26})$$

The term  $\langle \delta S_{i_1}^{\alpha_1} \delta S_{j_1}^{\beta_1} \delta S_{i_2}^{\alpha_2} \delta S_{j_2}^{\beta_2} \delta S_k^{\alpha_3} \rangle$  in Eq. (C25) can be dealt with as described previously in Appendix C 1. The outcome reads

$$\langle \delta S_{i_1}^{\alpha_1} \delta S_{j_1}^{\beta_1} \delta S_{i_2}^{\alpha_2} \delta S_{j_2}^{\beta_2} \delta S_k^{\alpha_3} \rangle = 4 \langle \delta S_{i_1}^{\alpha_1} \delta S_{i_1}^{\alpha_2} \rangle \langle \delta S_k^{\alpha_3} \delta S_k^{\beta_1} \delta S_k^{\beta_2} \rangle, \quad (\text{C27})$$

where the factor of 4 comes from the number of different ways of pairing site indices yielding non-vanishing results. On the other hand,

$$\langle U^2 \rangle = \frac{1}{2} \sum_{ij} J_{ij}^{\alpha_1 \beta_1} J_{ij}^{\alpha_2 \beta_2} \langle \delta S_i^{\alpha_1} \delta S_i^{\alpha_2} \rangle \langle \delta S_j^{\beta_1} \delta S_j^{\beta_2} \rangle, \quad (\text{C28})$$

and in turn

$$\frac{\partial \langle U^2 \rangle}{\partial m_k^{\alpha_3}} = \frac{\partial \lambda_k^{\alpha_3}}{\partial m_k^{\alpha_3}} \sum_{ij} J_{ij}^{\alpha_1 \beta_1} J_{ij}^{\alpha_2 \beta_2} \frac{\partial \langle \delta S_i^{\alpha_1} \delta S_i^{\alpha_2} \rangle}{\partial \lambda_k^{\alpha_3}} \langle \delta S_j^{\beta_1} \delta S_j^{\beta_2} \rangle, \quad (\text{C29})$$

where  $\frac{\partial \lambda_k^{\alpha_3}}{\partial m_k^{\alpha_3}} \simeq 3$ . Here, we ignored higher order terms which do not contribute to the degeneracy lifting terms of fourth or sixth order in components of  $\mathbf{m}$  in Eq. (C25). The  $\frac{\partial \langle \delta S_i^{\alpha_1} \delta S_i^{\alpha_2} \rangle}{\partial \lambda_k^{\alpha_3}}$  expression can be calculated using Eq. (C12). Substituting Eqs. (C27, C29) in Eq. (C25), we obtain the final expression which can be represented by a “fused” diagram shown in Fig. 8. This diagram has a new type of vertex represented with a red square. This vertex is labeled with only one site index and its mathematical expression is:

$$\langle \delta S_k^{\alpha_3} \delta S_k^{\beta_1} \delta S_k^{\beta_2} \rangle \langle \delta S_k^{\alpha_3} \delta S_k^{\gamma_1} \delta S_k^{\gamma_2} \rangle, \quad (\text{C30})$$

where again there is a sum over repeated Greek superscripts. As a result, the final expression reads:

$$\langle U^2 T_2 \rangle = -\frac{\partial \lambda_k^{\alpha_3}}{\partial m_k^{\alpha_3}} \sum_{ijk} J_{ik}^{\alpha_1 \beta_1} J_{ik}^{\alpha_2 \beta_2} J_{jk}^{\alpha_3 \beta_3} J_{jk}^{\alpha_4 \beta_4} \langle \delta S_i^{\alpha_1} \delta S_i^{\alpha_2} \rangle \langle \delta S_k^{\alpha_3} \delta S_k^{\beta_1} \delta S_k^{\beta_2} \rangle \langle \delta S_k^{\alpha_3} \delta S_k^{\beta_3} \delta S_k^{\beta_4} \rangle \langle \delta S_j^{\alpha_4} \delta S_j^{\beta_4} \rangle. \quad (\text{C31})$$

From this point on, one can adopt the procedure presented previously for the  $\langle U^n \rangle$  terms to obtain the final result. All other averages that contain  $T_n$  e.g.  $\langle U^{n_1} T_{m_1} T_{m_2} \rangle$  and  $\langle T_{n_1}^{m_1} T_{n_2}^{m_2} \rangle$  where  $n_i, m_i$  with  $i = 1, 2$  are natural numbers, can be calculated following a similar procedure.

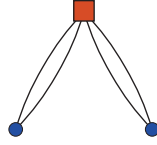


FIG. 8. “Fused” diagram corresponding to  $\langle U^2 T_2 \rangle$ . The vertex represented by the (red) square represents the following:  $\langle \delta S_k^{\alpha_3} \delta S_k^{\beta_1} \delta S_k^{\beta_2} \rangle \langle \delta S_k^{\alpha_3} \delta S_k^{\gamma_1} \delta S_k^{\gamma_2} \rangle$ .

#### 4. On Diagrams

In this subsection, we make a few comments about the different diagrams that appear in the E-TAP expansion.

First we focus on diagrams corresponding to  $\langle U^n \rangle$  with  $n = 2, 3, \dots$ , in Eq. (C5). The diagrams for  $n = 3, 4$  are illustrated in Fig. 7. In this figure, blue circles represent a given lattice site and the number of lines (bonds) connected to the circles is the number of paired  $\delta S$ 's at that lattice site. The degree of the vertices is indicated above each diagram and is written in the form of  $(\alpha_1, \dots, \alpha_m)$  where  $m$  is the number of vertices and  $\alpha_i$  is the number of bonds connected to vertex  $i$ . The number of times that each diagram appears in the process of pairing  $\delta S$ 's, referred to as diagram count, is indicated in parentheses to the right of each diagram in

Fig. 7. The diagrams are distinguished by the degree of their vertices and their adjacency matrix eigenvalues.<sup>59</sup> The adjacency matrix  $M$  is a  $m \times m$  matrix with matrix elements  $M_{ij}$  ( $0 < i, j \leq m$ ) corresponding to the number of lines connecting the  $i$ -th vertex to the  $j$ -th vertex. We enumerate these diagrams using a computer program in which we generate all possible diagrams given a certain number of vertices of a given degree. Then, we remove all diagrams that include any onsite interactions which are forbidden since there is no onsite interaction in the Hamiltonian of Eq. (1). For the remaining diagrams, we build their adjacency matrix which we then diagonalize.  $M$  is a symmetric matrix and thus has a set of real eigenvalues which defines a graph equivalence class. Graph with the same adjacency matrix eigenvalues correspond to graphs with the same topology, i.e. they are isomorphic.<sup>69</sup> The number of graphs with a given topology is noted in parentheses to the right of the graph in Fig. 7.

The fifth and sixth order terms of the E-TAP expansion in  $\beta$ , contain, respectively, 9 and 26 types of connected diagrams. We note that the contribution of disconnected diagrams in Eq. (C5) adds up to zero.<sup>59</sup>

The so-called fused diagrams appear in terms of the form  $\langle U^m T_n \rangle$  and  $\langle T_m T_n \rangle$ , where  $m, n \geq 2$  are positive integers. They are constructed by fusing the diagrams similar to the ones in Fig. 7 together. An example of such a fused diagram and the corresponding details of its definition is given in the caption of Fig 8.

We note that some of the diagrams do not cover lattice sites beyond one tetrahedron, for example the triangular diagram and the two site diagram in the top row of Fig. 7. Only, diagrams of this type contribute to fluctuations incorporated in the cluster mean-field theory (c-MFT) calculations. However, their effect is incorporated to all orders in  $\beta$  in the c-MFT calculation.

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- <sup>51</sup>  $\langle \dots \rangle$  represents a thermal Boltzmann average.
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